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| Title: | Using femtosecond pump-probe spectroscopy to shed new light on complex materials |
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| Issued: | 2021-09-20 |

# Using Femtosecond Pump-Probe Spectroscopy to Shed New Light on Complex Materials

Rohit Prasankumar

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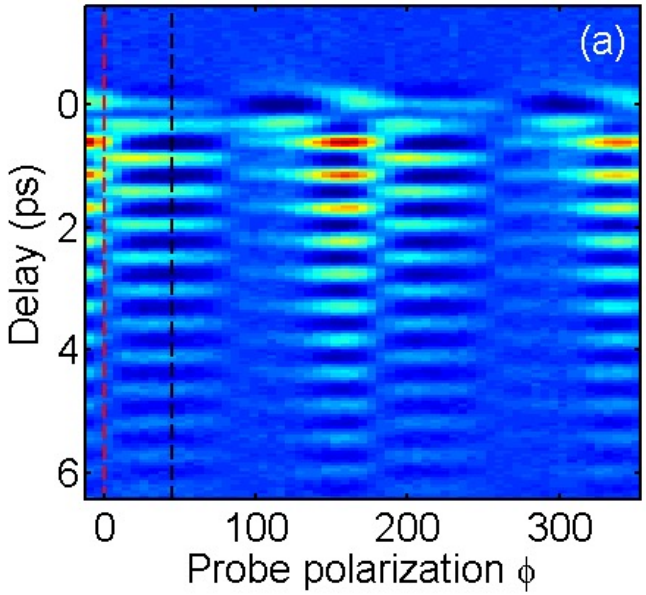
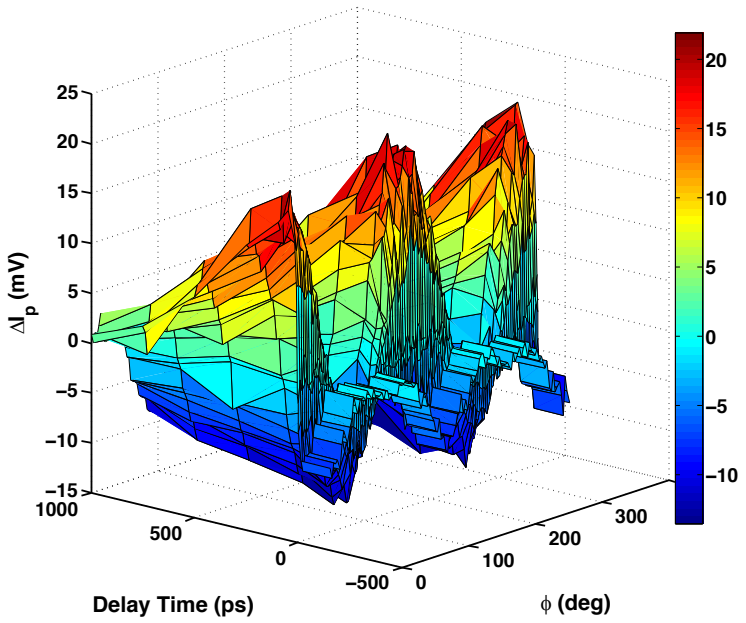
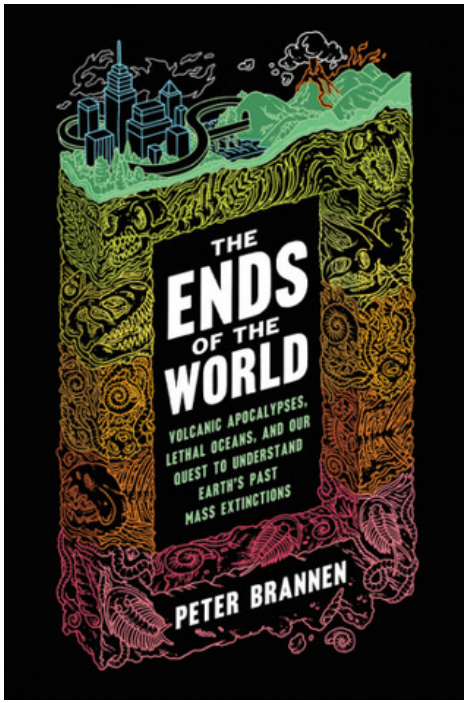
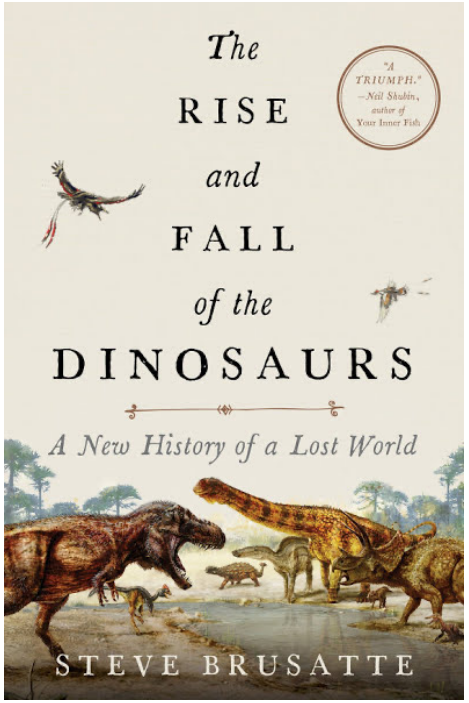



Los Alamos NATIONAL LABORATORY EST. 1943

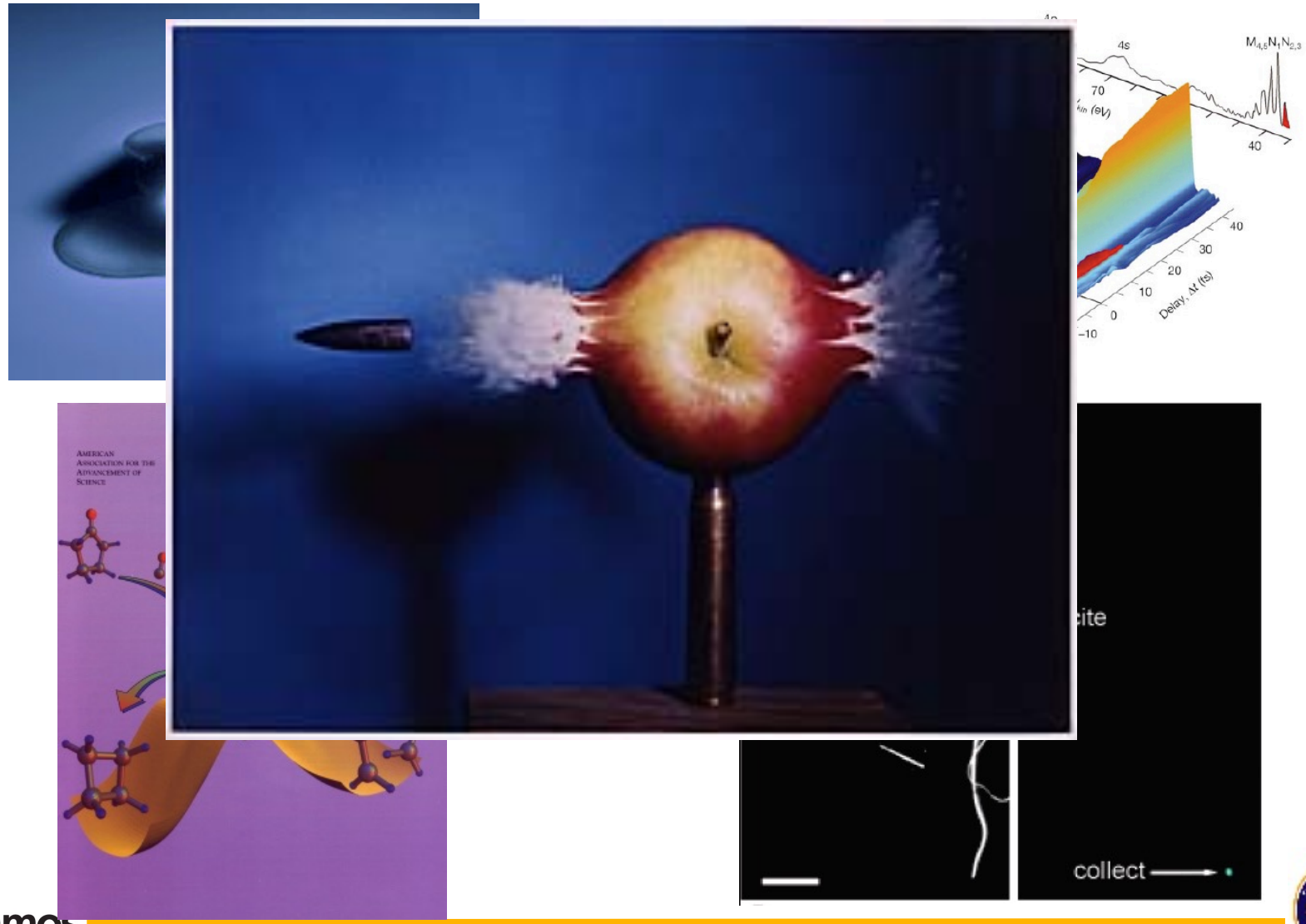
# Geological timescales




- We know a lot about events that have occurred during the earth's history, on timescales of millions of years
  - Five major extinctions ranging from ~450 to 65 million years ago
- We are much less familiar with events that occur on "ultra"-short timescales of less than a billionth of a second

# What kind of phenomena occur at ultrashort timescales?



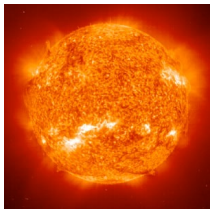
# A femtosecond ($10^{-15}$ s) is very short

## Ultrafast pulse duration: 10 fs

*In 10 femtoseconds:*

A passenger jet travels 2% of the width of one hydrogen atom.

The whole Earth receives 1.2 kJ (0.3 Cal) of energy from the sun.



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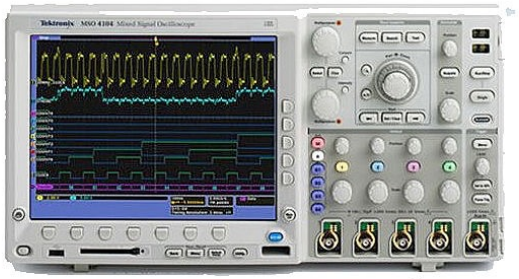
Interest on the U.S. national debt increases the total by $0.00000000013.

# Getting a better feel for ultrashort timescales

| **1 picosecond=$10^{-12}$ seconds** | **1 femtosecond=$10^{-15}$ seconds** |
|---|---|
| 1 year ⇨ 14 billion years | 71 picoseconds ⇨ 1 second |
| 1 day ⇨ 14 billion years | 183 femtoseconds ⇨ 1 second |
| 1 hour ⇨ 14 billion years | 7.6 femtoseconds ⇨ 1 second |

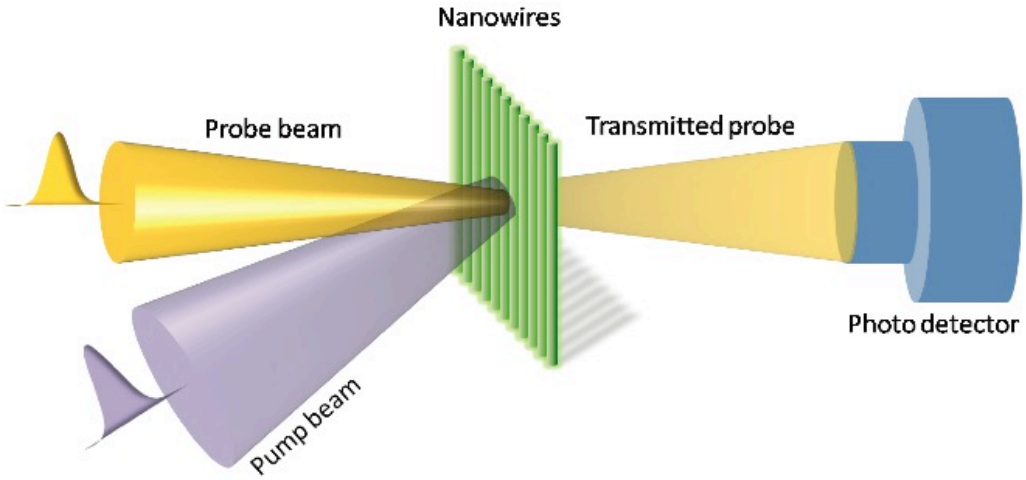
Unlike in geology, we can actually do experiments at ultrashort timescales, so we should be able to understand the "ultrafast" world much better!

# How can we measure phenomena occurring on an ultrafast time scale?



Conventionally, one would use a fast oscilloscope to do this…
but what can you do if even the fastest oscilloscope is too slow?

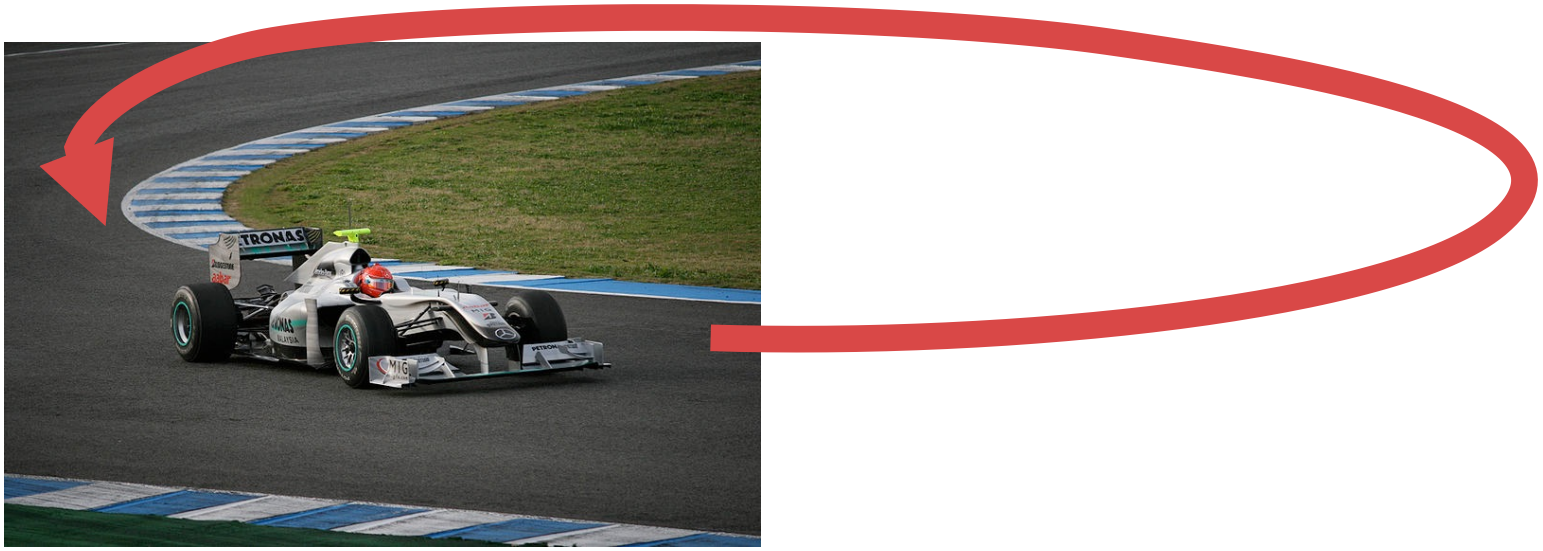
Ultrafast optical spectroscopy is the only technique capable of measuring processes at the fundamental time scales of electron, lattice and nuclear motion



# Time domain measurements: an analogy

Imagine you want to make a movie of an extremely fast car as it drives past you...

**Problem:** What if the car passes by so quickly that your camera can only capture *one frame* before the car is gone?



**Solution:** Take a picture of the car at a slightly different position on each lap (assuming it follows the same path each lap)... Then combine all the photos into a movie.

# Freezing motion with ultrashort light pulses

# Why do we need to understand ultrafast phenomena?

- Fundamental scientific interest:
  - How fast are different excitations created and destroyed in complex systems?
  - Can we track various processes through which excitations lose energy as they return to equilibrium in both space and time?
  - How do different subsystems (electrons, lattice, spins) interact and exchange energy on ultrashort timescales?
  - Can we extract intrinsic material properties from these measurements?

- Application relevance:
  - Energy-related applications:
    - Charge separation in solar cells
    - Optimizing solid-state lighting devices (e.g., LEDs)
    - Coupled heat and electrical transport in thermoelectrics
  - Ultimate speed limitations of hard drives, semiconductor lasers, switches, etc.
  - Using ultrashort laser pulses to *control* complex phenomena
    - Photoinduced phase transitions

**How do we generate and use femtosecond optical pulses for studying ultrafast phenomena?**

# Outline

**We use femtosecond pump-probe spectroscopy to study complex materials, revealing new information about their properties that can impact future applications**

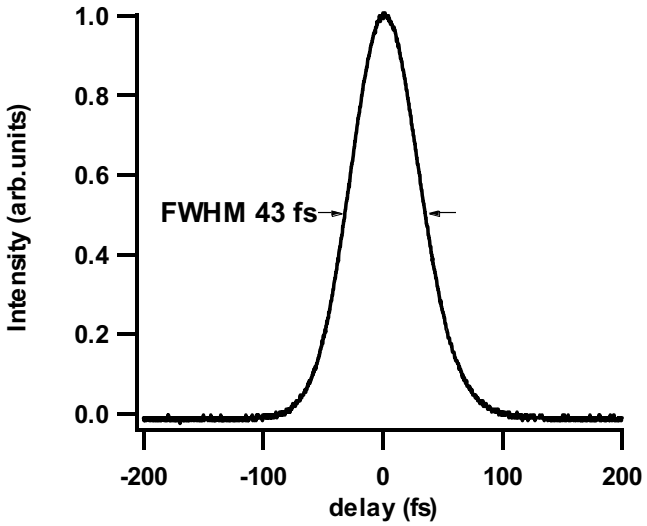
- Background on ultrashort pulse generation and pump-probe spectroscopy
- Examples of carrier relaxation in semiconductor nanostructures:
  - Ultrafast carrier relaxation across multiple spatial dimensions in a semiconductor quantum dots-in-a-well (DWELL) heterostructure
    - Can we track carrier relaxation from 3D to 2D to 0D in a novel heterostructure?
  - Time-resolved carrier dynamics in ensemble and individual quasi-1D semiconductor nanowires
    - What pathways do photoexcited carriers take as they lose energy in a quasi-1D system?
- Going beyond simple semiconductors: using ultrashort light pulses to probe and even control more complex materials

# Ultrashort pulse basics

- Typical ultrashort pulse has a sech$^2$ or Gaussian intensity profile:

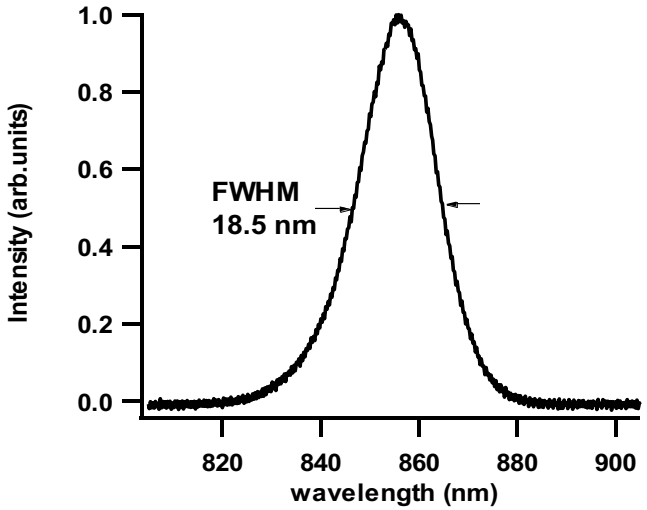
$$I \sim Ae^{-\frac{t^2}{\tau^2}}$$

where $\tau$ is the pulsewidth.




- The temporal profile of the pulse is related to its spectral shape through Fourier transforms

- Both the temporal and spectral shape of the pulse can be measured experimentally

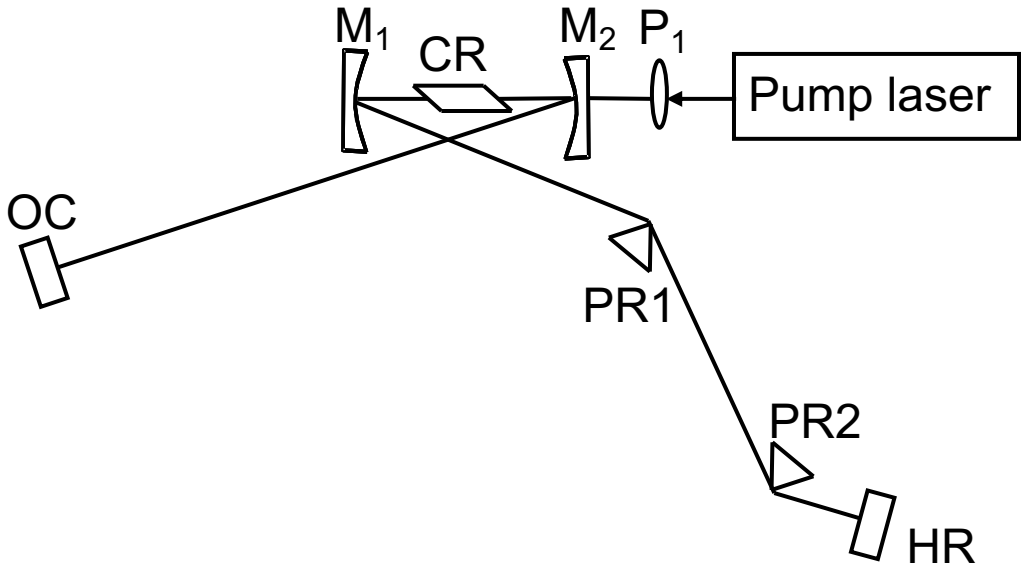


# Ultrashort pulse generation

- How is an ultrashort pulse generated?
  - Need a mechanism that favors pulsing over continuous wave (cw) operation in a laser cavity
  - Need to minimize dispersion (variation of the index of refraction with frequency) to prevent pulse from spreading out in time

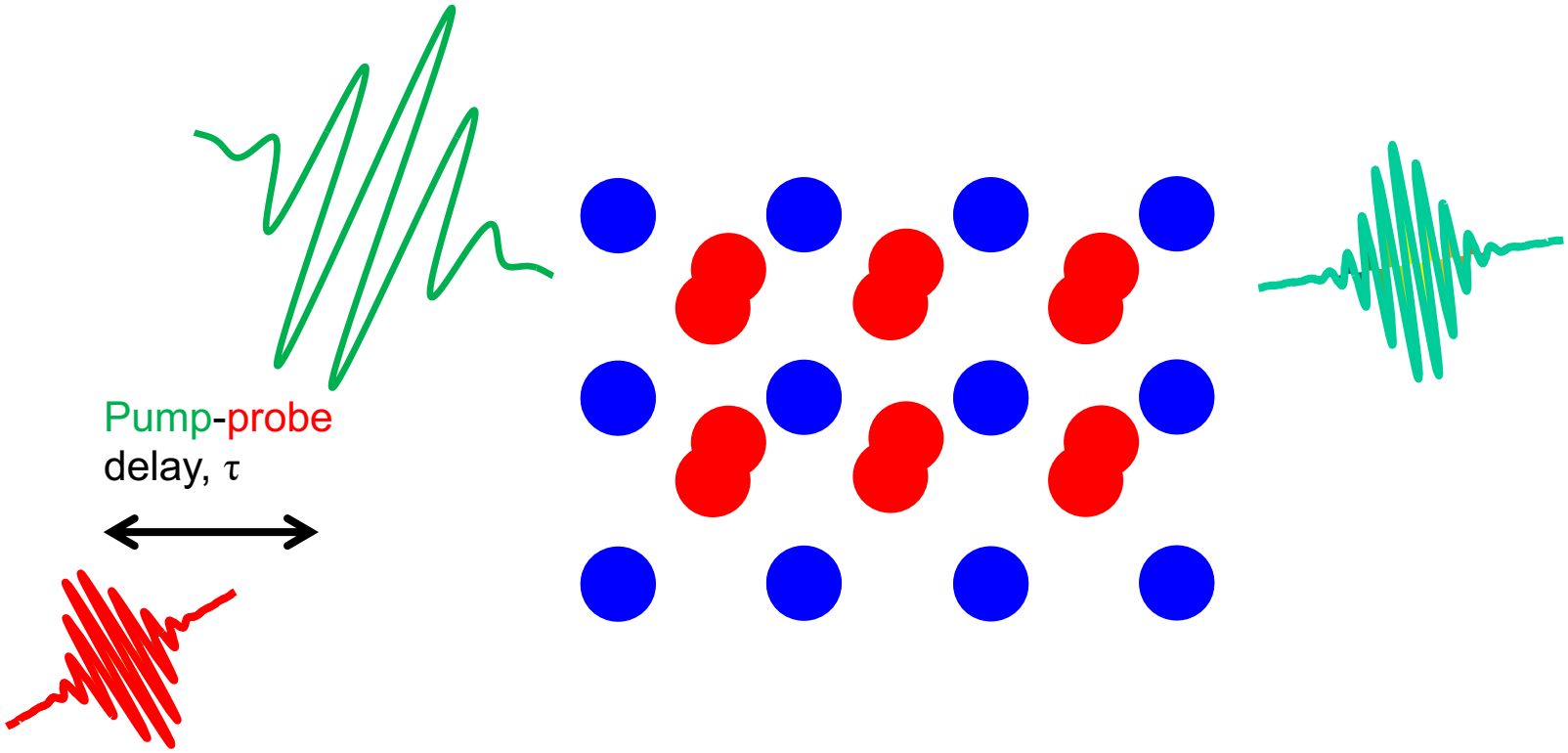
- Passive mode-locking
  - "Mode-locking"-frequency domain term
  - Uses an intensity dependent loss or gain in the laser cavity to favor pulsed operation
  - Prisms or other dispersive elements are introduced into the cavity to shape the pulse-"soliton-like" pulse shaping

# Ultrafast laser design




- Ti:$Al_2O_3$ is the most common material for ultrafast lasers
  - 800 nm wavelength (1.55 eV energy)
  - Easily pumped with an argon or solid-state laser
  - Stable, broad bandwidth solid-state source of short pulses
- Standard four mirror cavity design
- Prisms used for dispersion compensation

# Femtosecond pump-probe spectroscopy

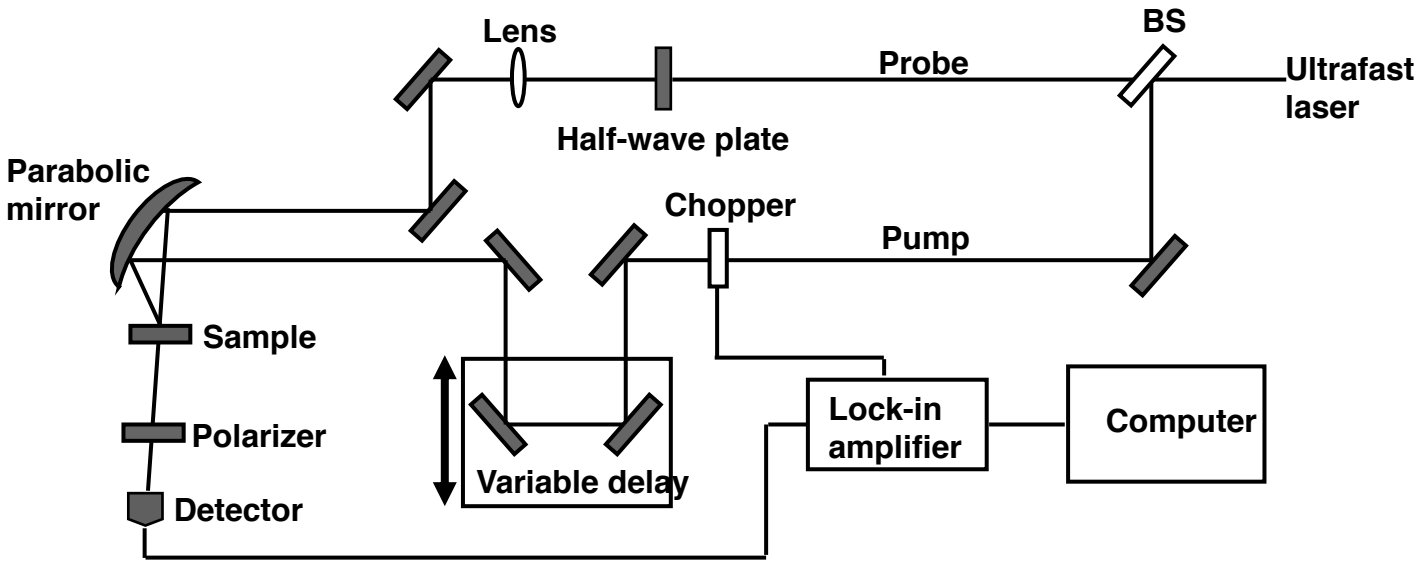



A pump pulse changes the material, and the probe pulse monitors these ultrafast changes.



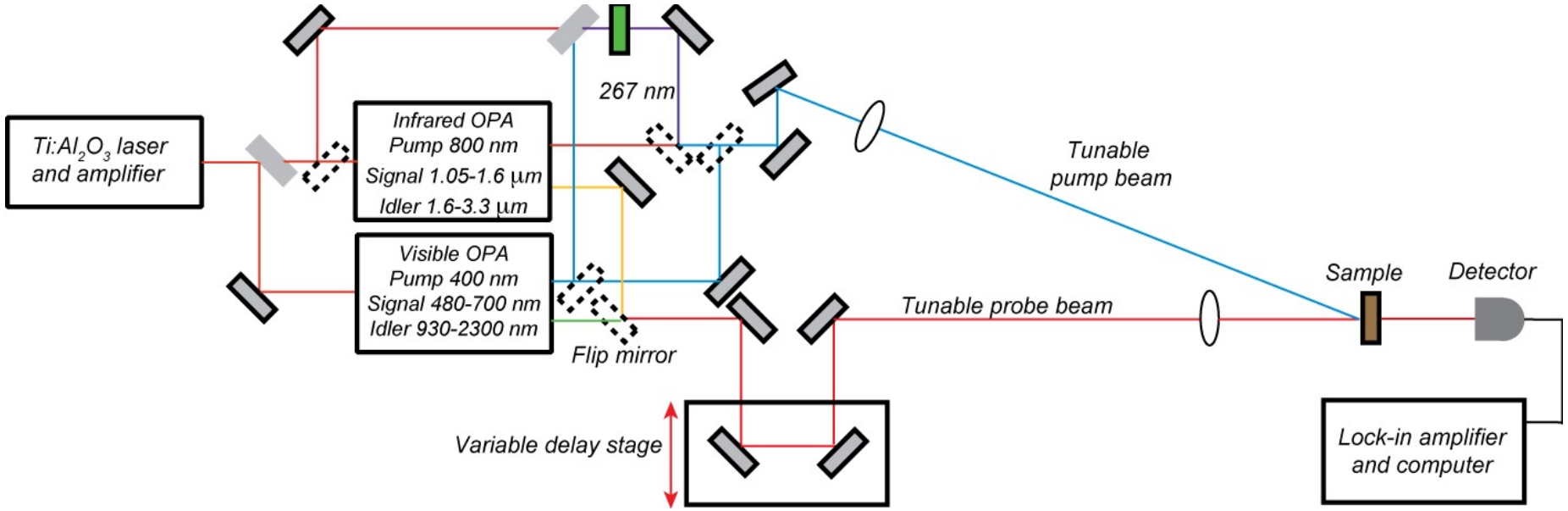
R. P. Prasankumar and A. J. Taylor (eds.), *Optical Techniques for Solid-State Materials Characterization* (CRC Press, 2011)

# Basic degenerate pump-probe setup




- Pulses from the femtosecond laser are split by a beam splitter (BS) into pump (>90% of power) and probe (<10% of power) pulses
  - "Degenerate" experiment where pump and probe have the same wavelengths
- Movement of delay stage adjusts timing between pump and probe
- Parabolic mirror or lens focuses beams onto sample
  - Pump spot size larger than probe spot size
  - Probe intensity is measured at detector

# Broadband femtosecond time-resolved spectroscopy system



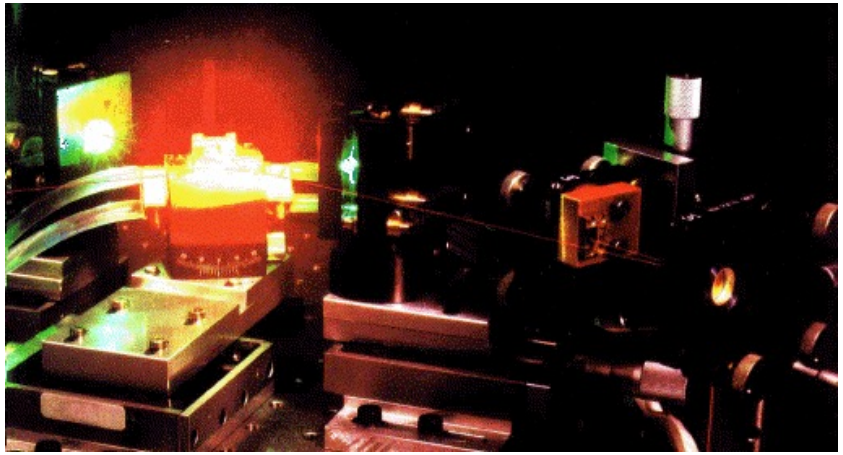

- Amplified $Ti:Al_2O_3$ laser system
  - 100 kHz repetition rate, 50 fs pulse duration, 10 μJ pulse energy at 800 nm
- Independently tunable pump and probe wavelengths from 400 nm-3.3 μm
- Flexible setup capable of studying dynamics in almost any physical, chemical, or biological system



R. P. Prasankumar, P. C. Upadhya and A. J. Taylor, Physica Status Solidi(b) (2009)

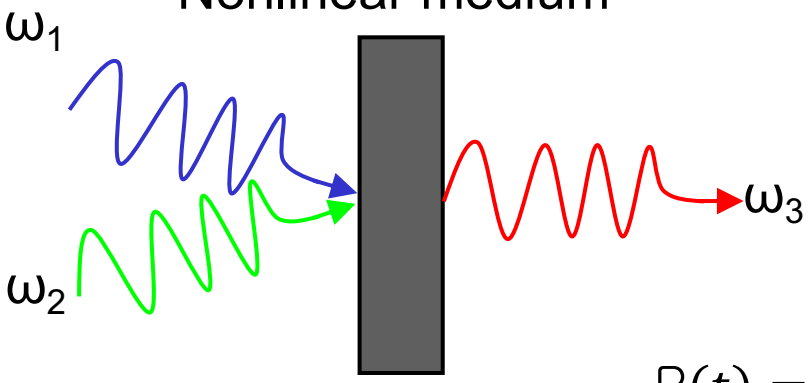
# Generating other frequencies with nonlinear optics

Femtosecond Ti:Sapphire laser



$Ti:Al_2O_3$ (Ti:sapphire) lasers/amplifiers can generate intense, ultrashort pulses with durations down to 4.5 fs, with wavelengths from 700-1000 nm.

Nonlinear medium




$\omega_3=\omega_1-\omega_2$

$\omega_3=\omega_1+\omega_2$

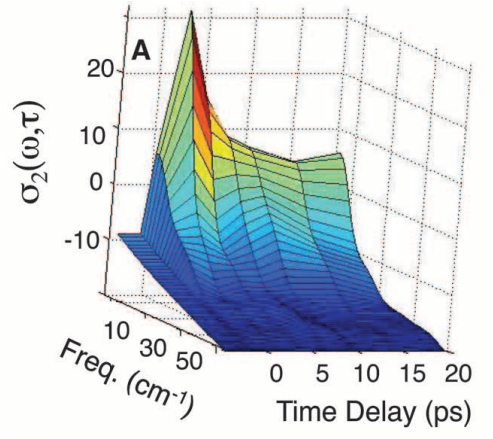
With nonlinear optics nearly any other frequency can be generated.

$$P(t) = \varepsilon_0\left(\chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + \cdots\right)$$

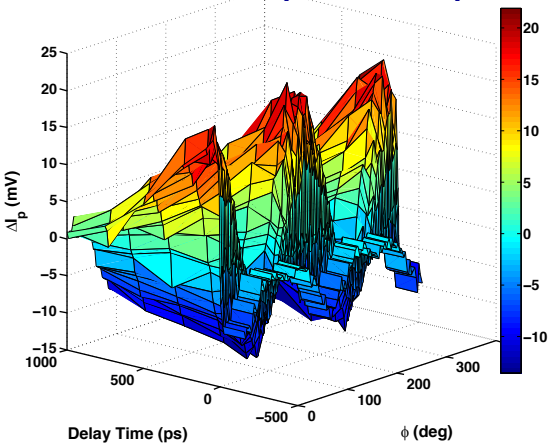
# More sophisticated ultrafast optical techniques

## Time-resolved terahertz (THz) spectroscopy

D. Fausti et al, *Science* (2011)

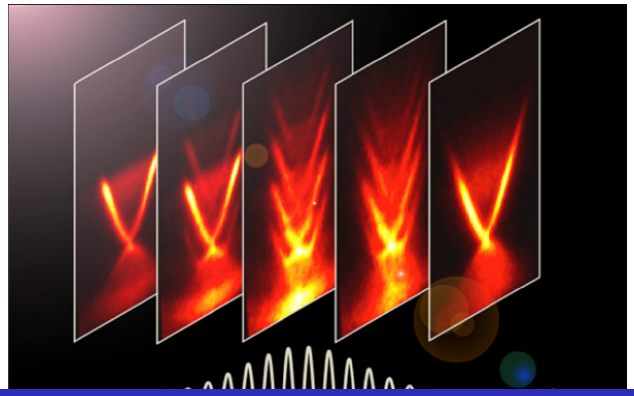



## Time-resolved second harmonic generation (TR-SHG)

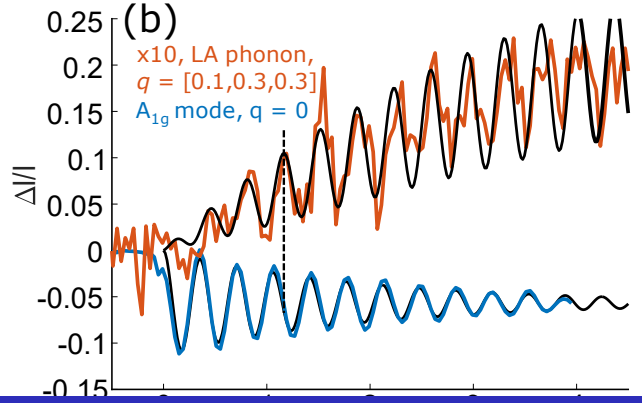



Y.-M. Sheu et al, *Nature Comm.* (2014)

## Time-and-angle-resolved photoemission (TR-ARPES)

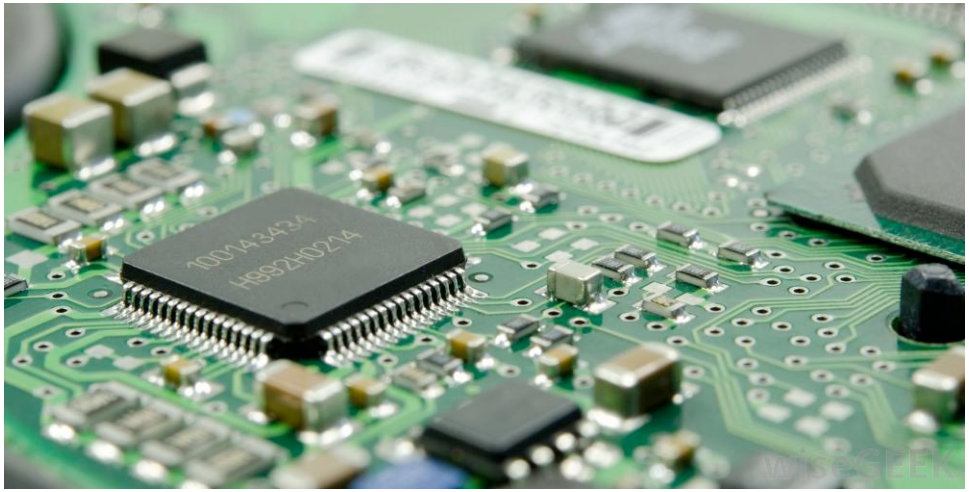


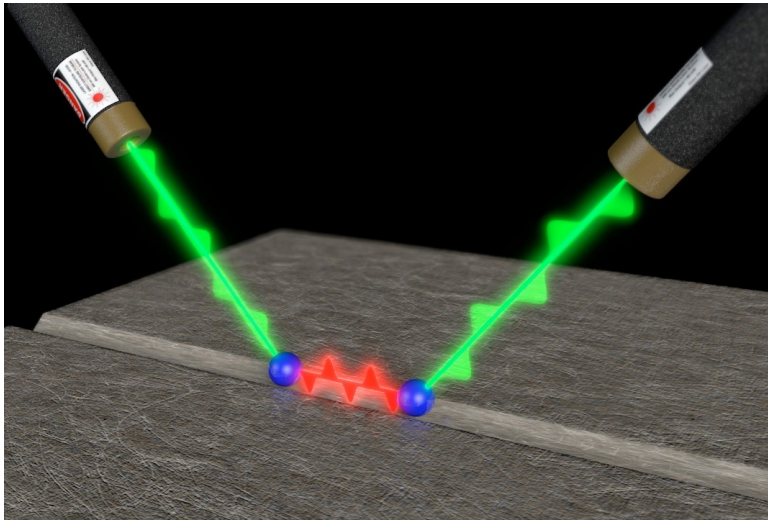
## Time-resolved X-ray diffraction




**These techniques all provide a greater degree of selectivity and specificity, enabling us to *directly* probe material properties**

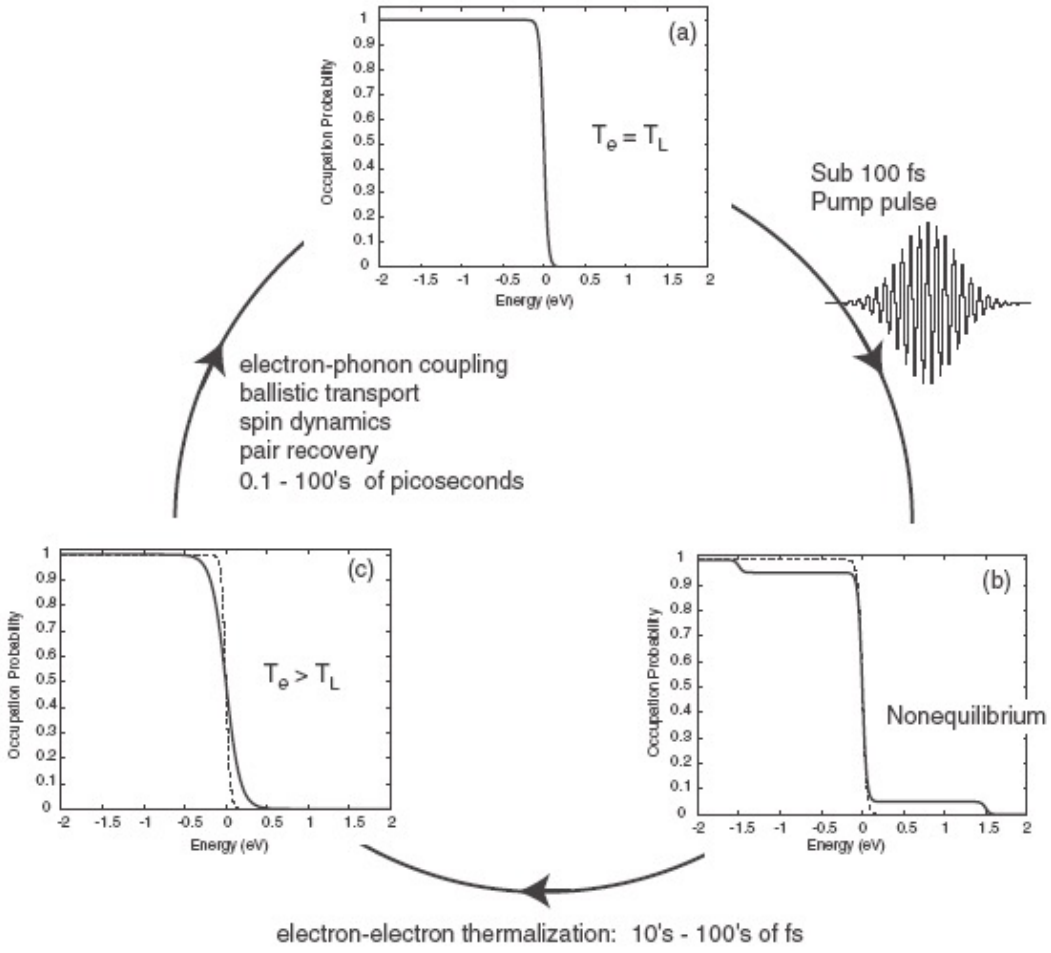
# Using ultrashort pulses to study conventional materials

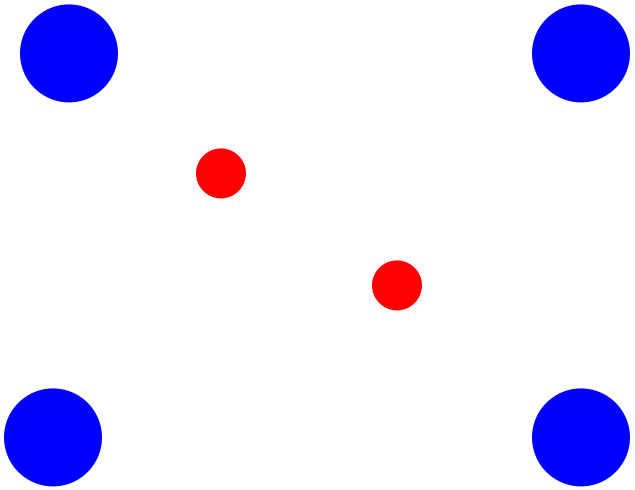




Conventional solids, such as semiconductors, metals, and insulators, have found a wide range of applications in modern technology

# Ultrafast dynamics in metals



$T_e = T_l$

$T_e > T_l$

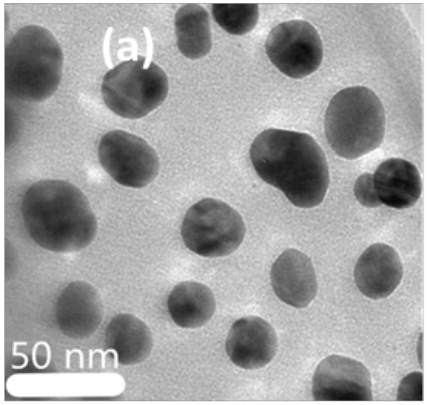
$T_e \rightarrow T_l$

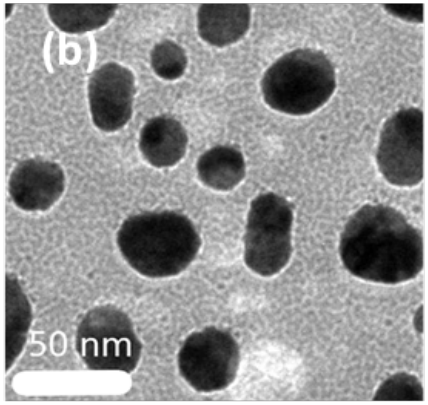
$T_e = T_l$

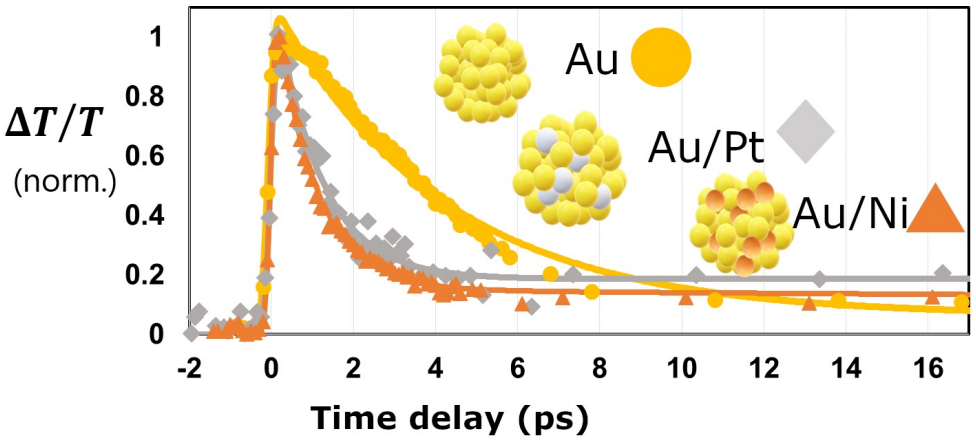


R. P. Prasankumar and A. J. Taylor (eds.), *Optical Techniques for Solid-State Materials Characterization* (CRC Press, 2011)

# Ultrafast dynamics in metal nanoparticles



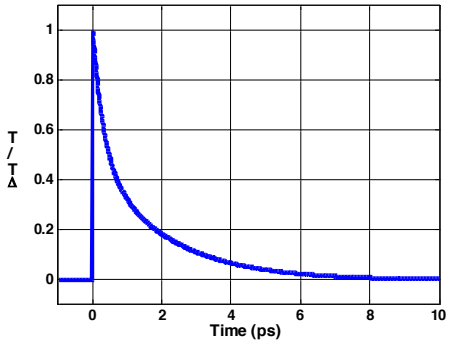



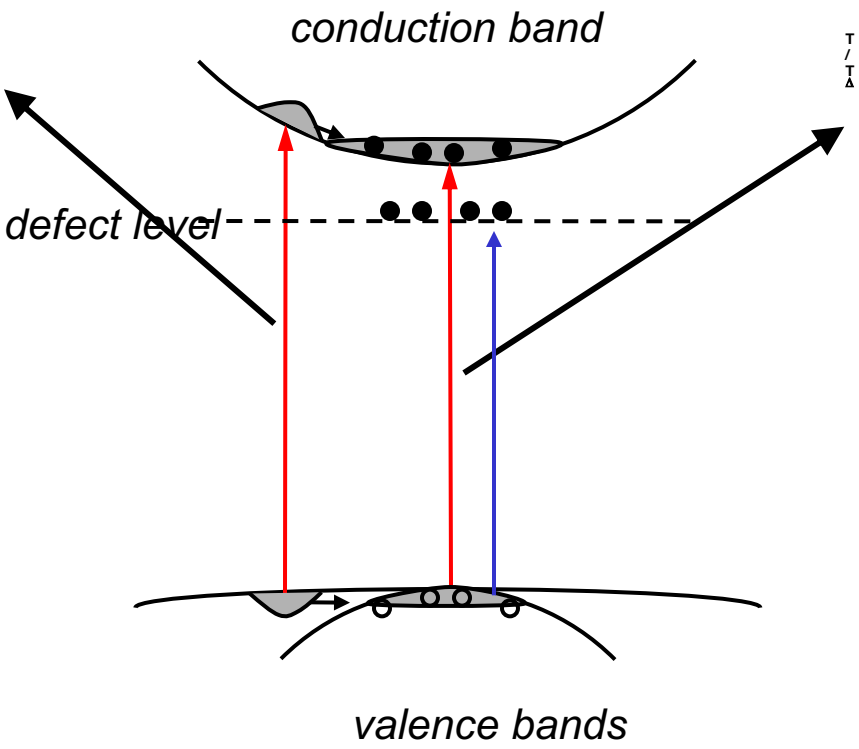

- (a) Au, (b) Au/Ni, and Au/Pt (not shown) nanoparticles were fabricated by our collaborators at New Mexico Tech
- Femtosecond pump-probe measurements demonstrated that carrier relaxation is longer in Au nanoparticles and faster in Pt and Ni coated nanoparticles
  - Occurs because photoexcited electrons relax quickly via interactions with phonons
  - Electron-phonon coupling is much stronger in Pt and Ni than Au
- Applications in photocatalysis
  - These nanoparticles are used to enhance chemical reactions, and the efficiency is influenced by carrier relaxation dynamics

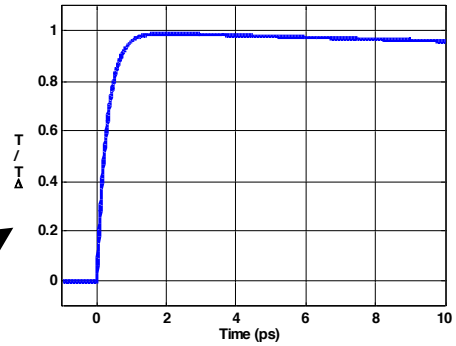


S. Sim et al, *Nanoscale* **12**, 10284 (2020)

# Carrier relaxation in bulk semiconductors





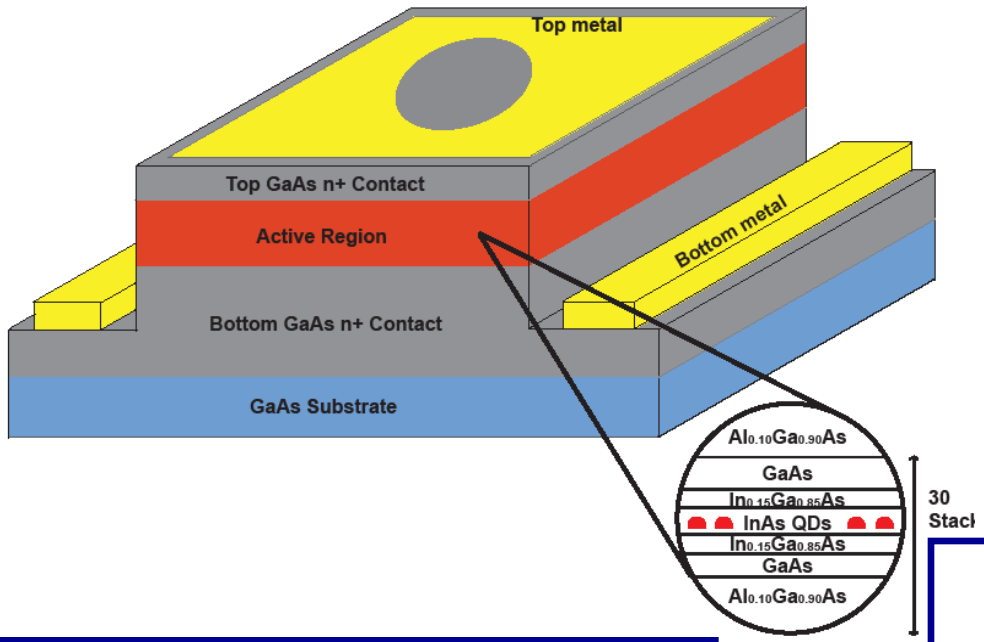

R. P. Prasankumar and A. J. Taylor (eds.), *Optical Techniques for Solid-State Materials Characterization* (CRC Press, 2011)

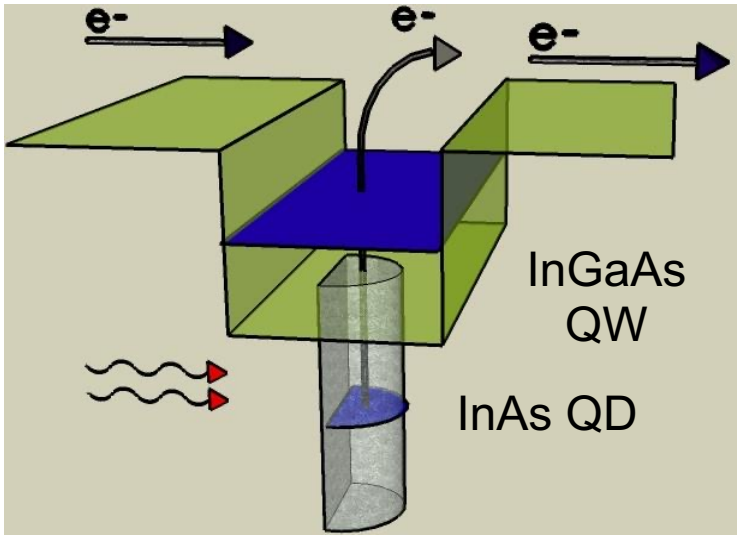
# Outline

**We use femtosecond pump-probe spectroscopy to study complex materials, revealing new information about their properties that can impact future applications**

- Background on ultrashort pulse generation and pump-probe spectroscopy
- Examples:
  - **Ultrafast carrier relaxation across multiple spatial dimensions in a semiconductor quantum dots-in-a-well (DWELL) heterostructure**
    - **Can we track carrier relaxation from 3D to 2D to 0D in a novel heterostructure?**
  - Time-resolved carrier dynamics in ensemble and individual semiconductor nanowires
    - What pathways do photoexcited carriers take as they lose energy in a quasi-1D system?
- Going beyond simple semiconductors: using ultrashort light pulses to probe and even control more complex materials

# Quantum dots in-a-well (DWELL) structures



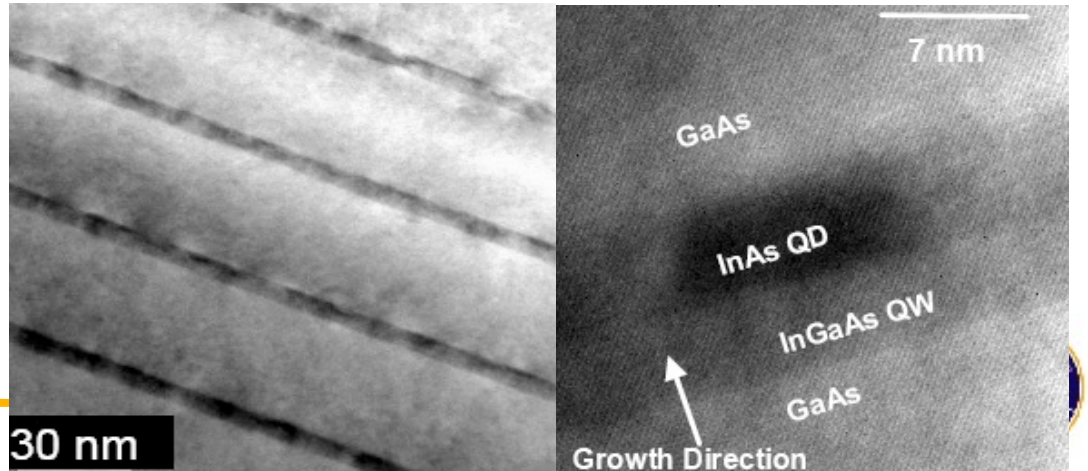




•InAs Quantum Dots are placed in an InGaAs Quantum Well

## Advantages of DWELL

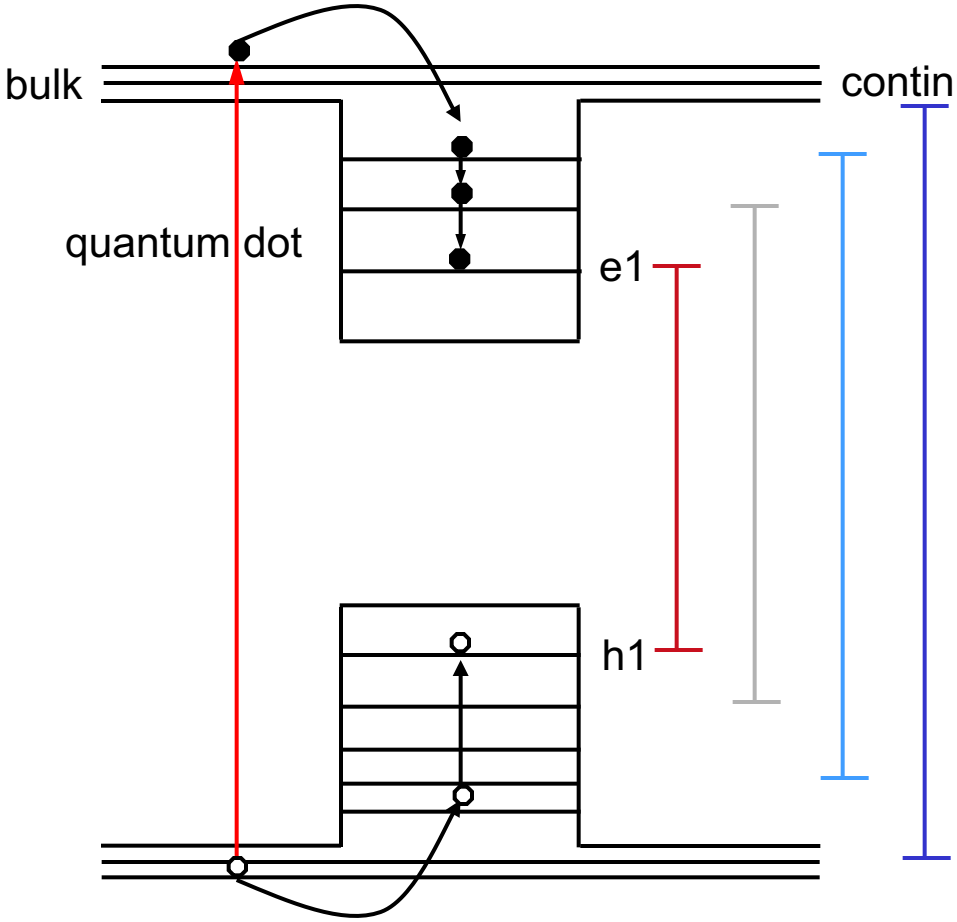
- Superior control of wavelength
- Better optical quality of QDs
- Reduction in dark current

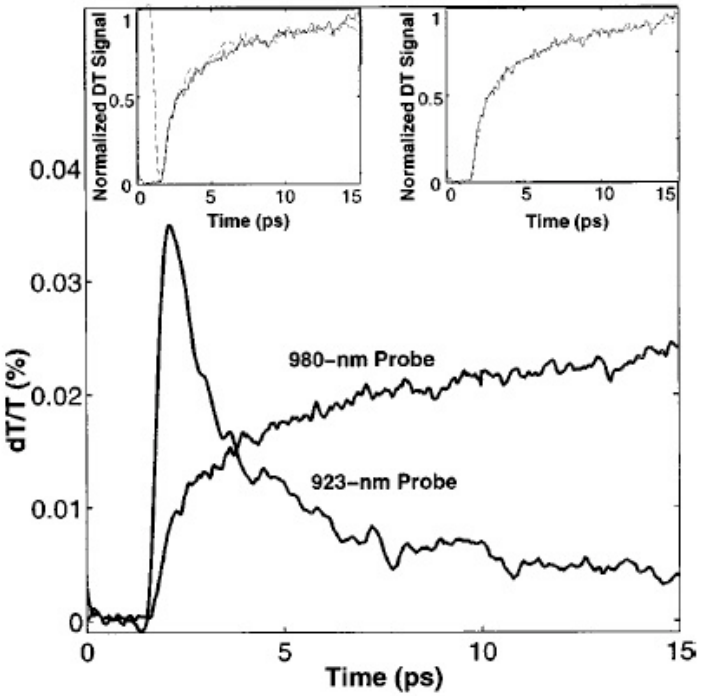
***HRTEM Image of a Single InAs QD***




S. Krishna, *Journal of Physics D (Applied Physics)* (2005)

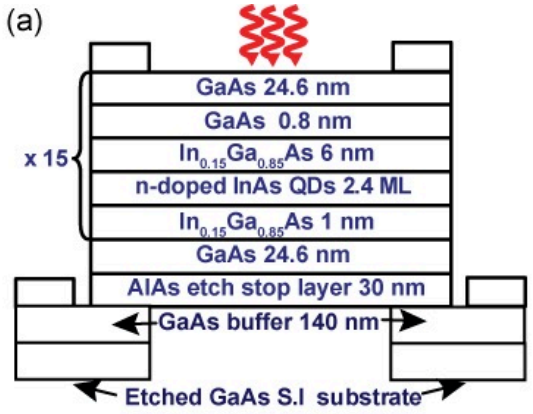
# Optical processes in quantum dots

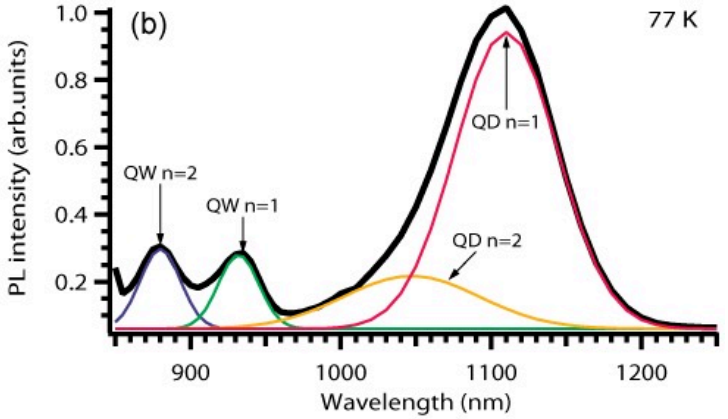


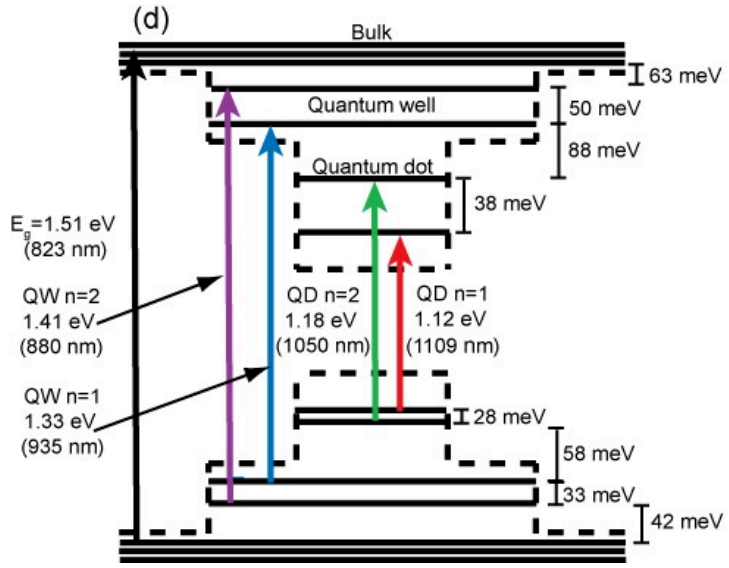



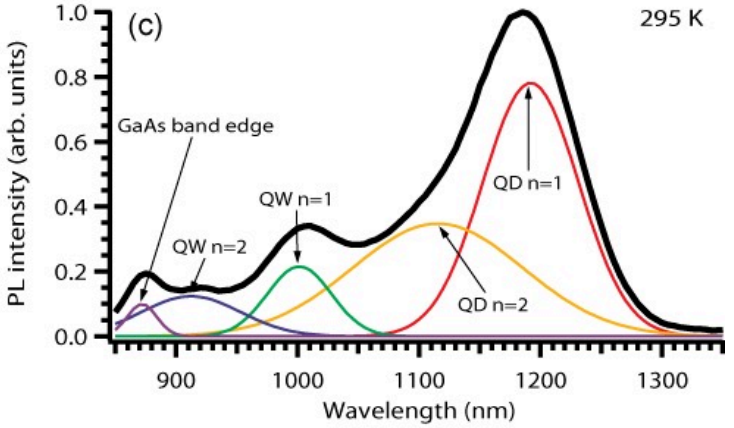

T. S. Sosnowski et al, *Phys. Rev. B* (1998)

# DWELL structure and energy levels



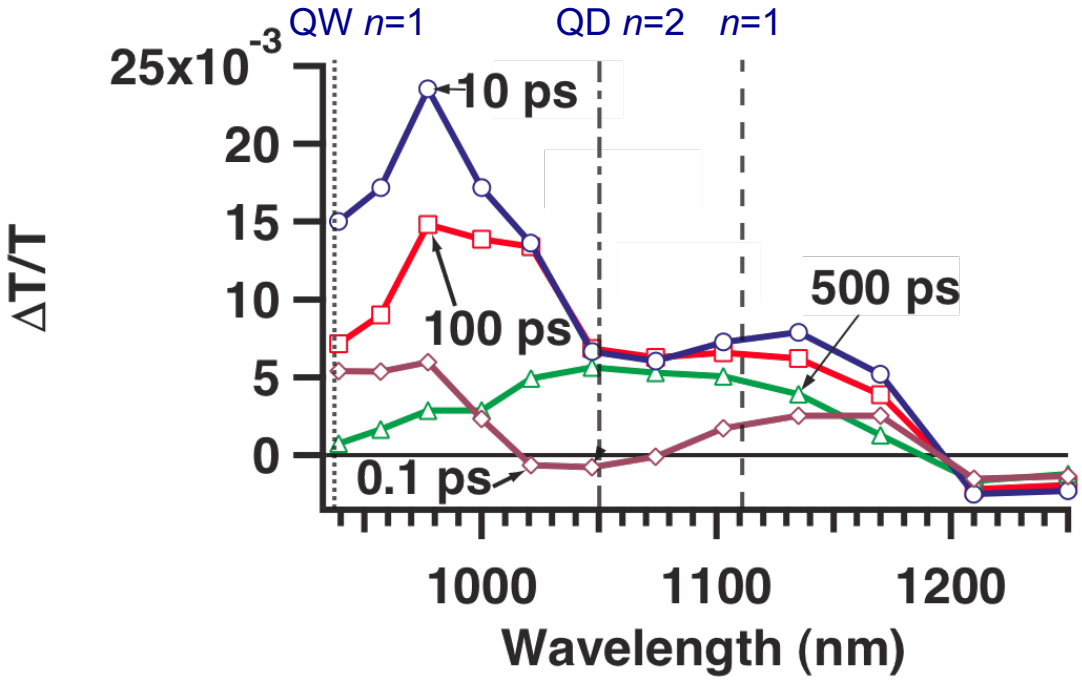




- Intensity-dependent photoluminescence (PL) measurements at 77 K and 295 K enable us to extract the DWELL energy level structure



S. Raghavan et al, *Appl. Phys. Lett.* (2002); R. P. Prasankumar et al, *Optics Express* (2008)

# Low temperature, low density DWELL differential transmission spectrum

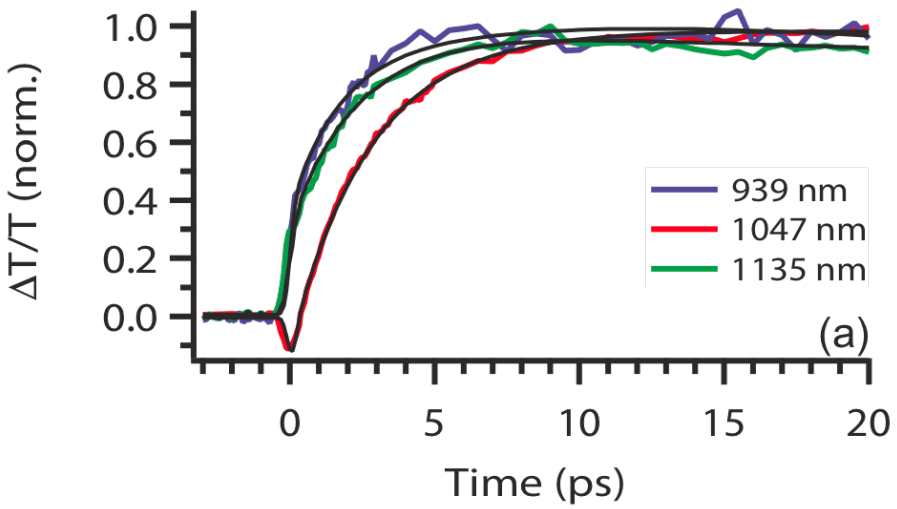


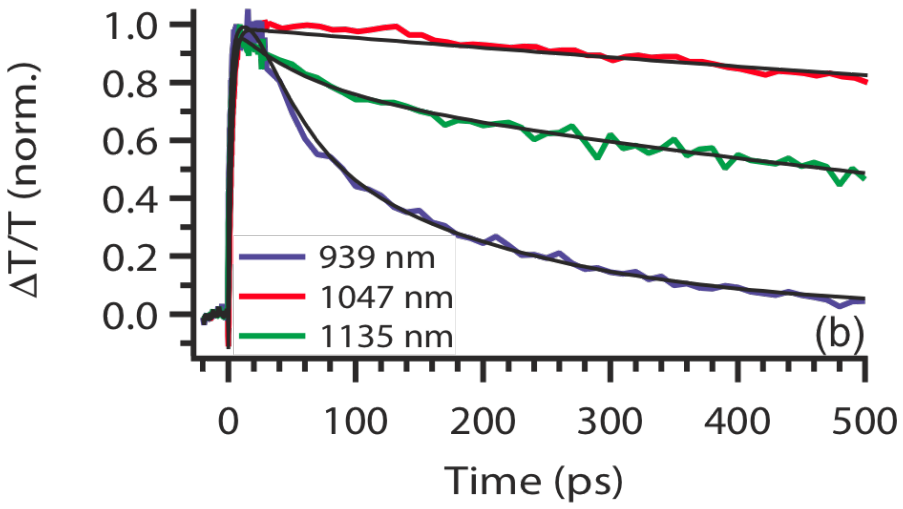

- 800 nm (1.55 eV) pump creates carriers in bulk layers at T=30 K
  - Pump fluence ~2.8 μJ/cm$^2$ excites ~4 electron-hole pairs (ehp)/dot
- "Camel back" shaped differential transmission spectrum (DTS) with peaks near QW *n*=1 state and *n*=1 QD state, long-lived relaxation at QD *n*=2 state
  - DT signal is proportional to number of electrons and holes in energy levels



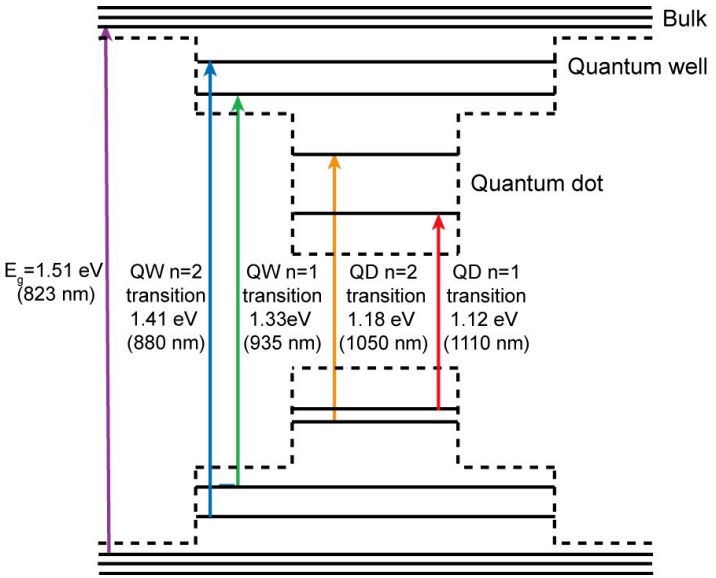
R. P. Prasankumar et al, *Optics Express* **16**, 1165 (2008)

# Time-resolved low temperature dynamics




- Rise times (2-4 ps) are comparable across the measured wavelength range
  - Rapid carrier capture into QWs
  - Auger-type electron-electron, electron-hole scattering fills QD states

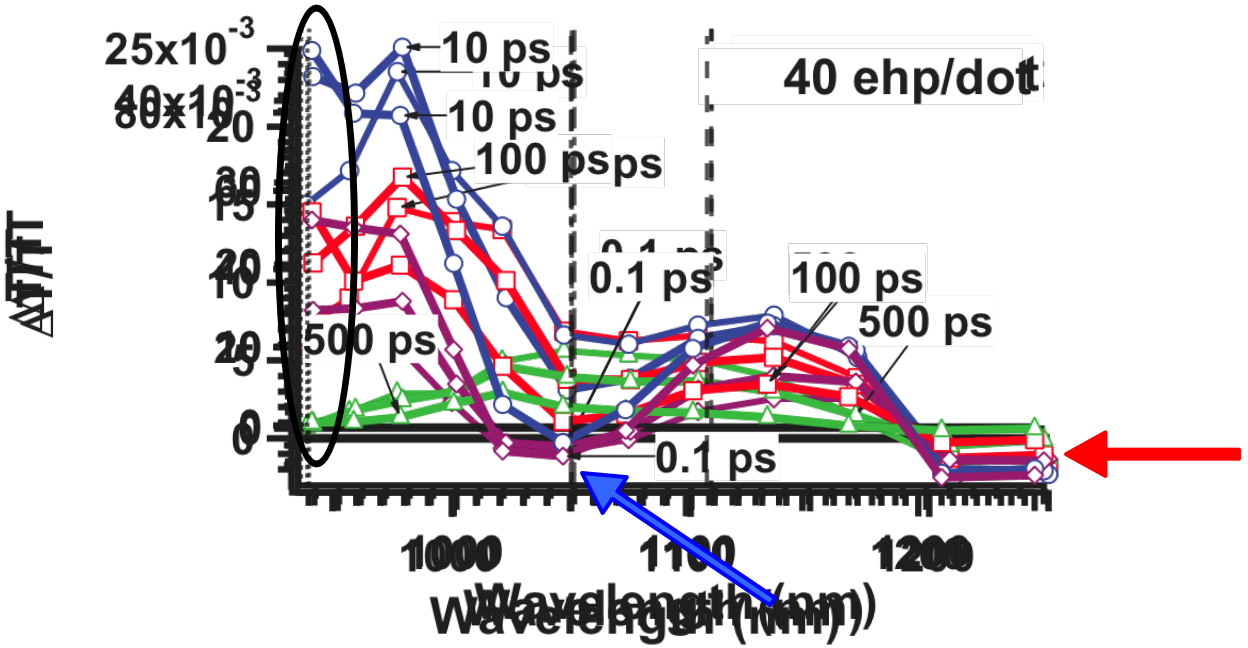



- Dynamics at long times
  - QD *n*=1 relaxation due to *e-h* recombination
  - State filling causes long-lived DT signal at QD *n*=2 level
  - Long-lived signal at QW state may be due to *e-h* scattering



R. P. Prasankumar et al, *Optics Express* **16**, 1165 (2008)

# Density-dependent differential transmission spectra

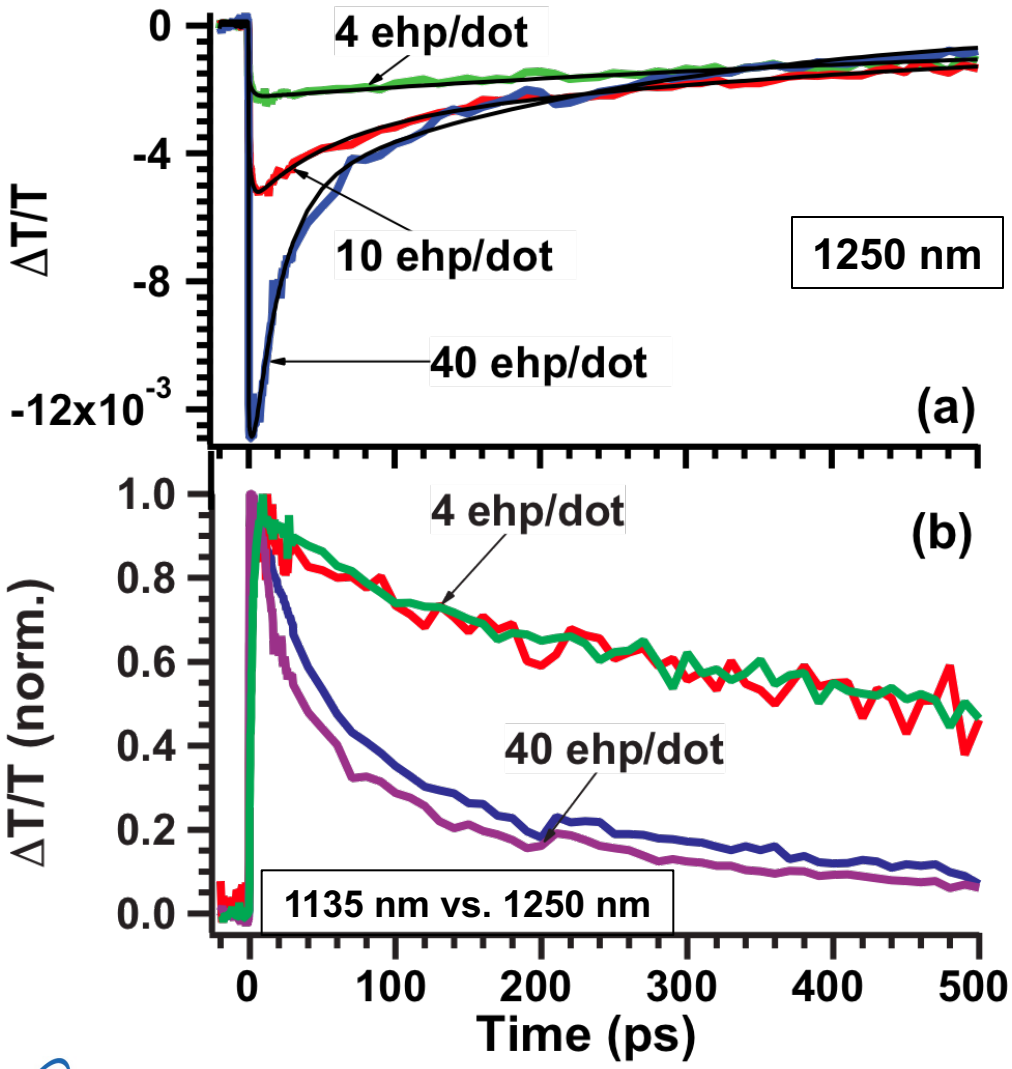



- Examine density dependence of DWELL carrier dynamics at T=30 K
- Negative DT signals observed at long wavelengths ($\lambda$>1200 nm) and from 1021-1074 nm
  - Typically attributed to Coulomb interactions between photoexcited carriers*



V. I. Klimov, *J. Phys. Chem. B* (2000)

# Interaction-induced ground state shift

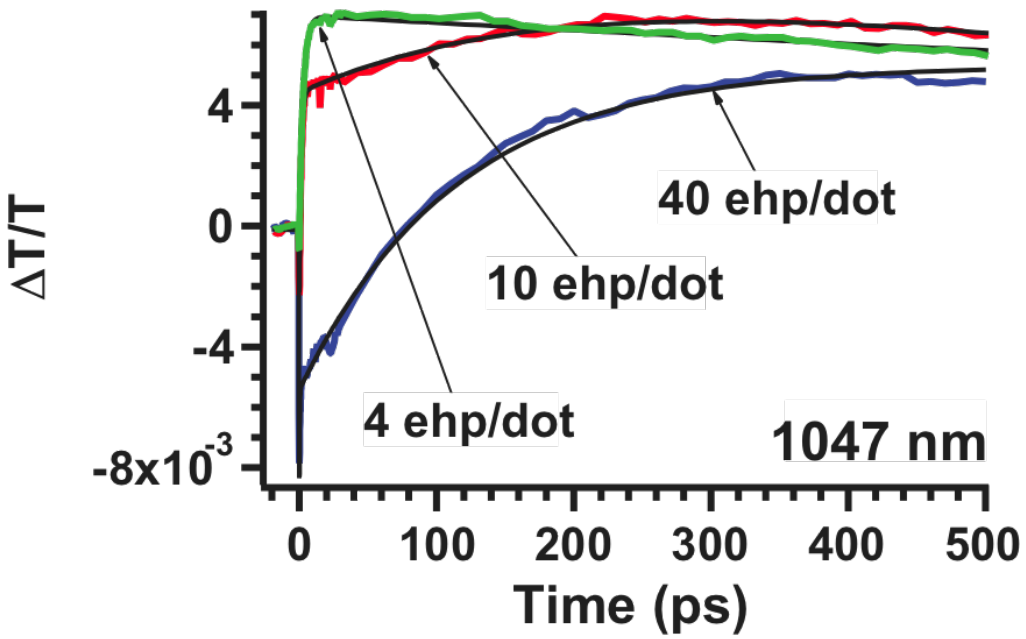



- Long wavelength DT signal remains negative at long times and with increasing carrier density
- Dynamics match that of QD ground state at all measured carrier densities
- This signal is due to a Coulomb interaction-induced red shift of the QD ground state
  - Simple model supports this



R. P. Prasankumar et al, *Appl. Phys. Lett.* **96**, 031110 (2010)

# Carrier dynamics at QD excited state

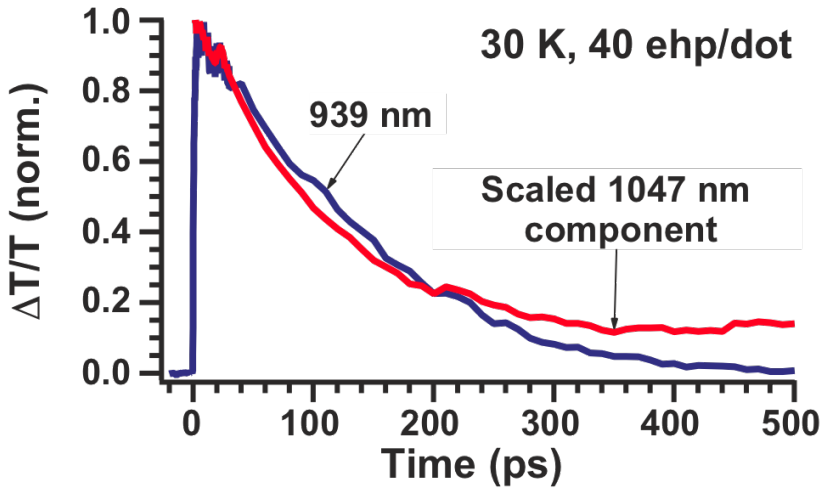



- Relaxation at QD excited state has three components:
  - Sub-ps negative signal due to Coulomb interaction-induced shifts of the QD *n*=2 state
  - Long-lived positive signal due to QD *n*=2 state filling
  - Additional induced absorption signal that substantially increases with carrier density
- QDs are filled at 6 ehp/dot, implying that this anomalous induced absorption signal is linked to the QW population



R. P. Prasankumar et al, *Appl. Phys. Lett*. **96**, 031110 (2010)

# Dynamic light-matter coupling at QD excited state




- Induced absorption signal and QW *n*=1 state signal are nearly identical, strongly suggesting that density-dependent induced absorption at QD *n*=2 state is linked to the QW population
- Possible mechanisms include:
  - Carrier-induced shifts of QW band edge
  - QD level shifts induced by 2D carrier density
  - Excitation-induced dephasing
- **This phenomenon is unique to the DWELL system and was not previously**

**Our studies demonstrate that ultrafast pump-probe can resolve carrier relaxation from 3D to 2D to 0D in semiconductor nanostructures**

# Outline

**We use femtosecond pump-probe spectroscopy to study complex materials, revealing new information about their properties that can impact future applications**

- Background on ultrashort pulse generation and pump-probe spectroscopy
- Examples:
  - Ultrafast carrier relaxation across multiple spatial dimensions in a semiconductor quantum dots-in-a-well (DWELL) heterostructure
    - Can we track carrier relaxation from 3D to 2D to 0D in a novel heterostructure?
  - **Time-resolved carrier dynamics in ensemble and individual semiconductor nanowires**
    - **What pathways do photoexcited carriers take as they lose energy in a quasi-1D system?**
- Going beyond simple semiconductors: using ultrashort light pulses to probe and even control more complex materials

# Carrier diffusion in one dimension

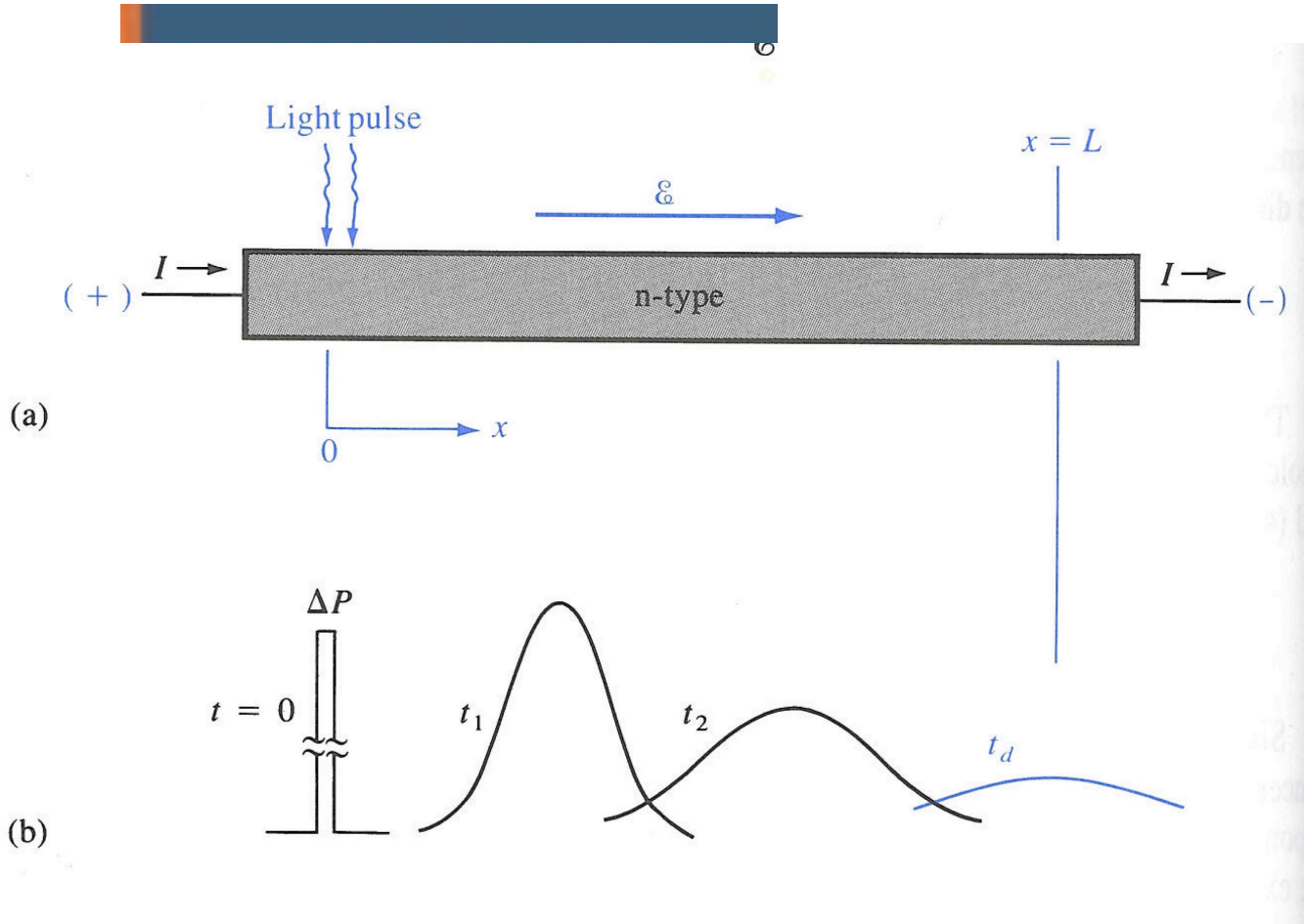
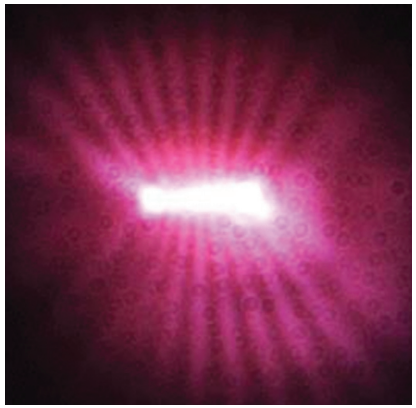


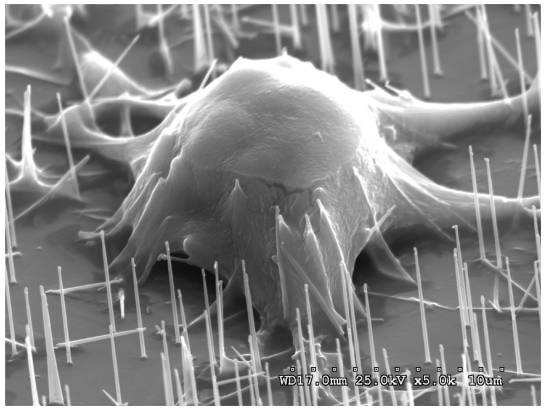

**Figure 4-18**
Drift and diffusion of a hole pulse in an n-type bar: (a) sample geometry; (b) position and shape of the pulse for several times during its drift down the bar.

# Semiconductor nanowires

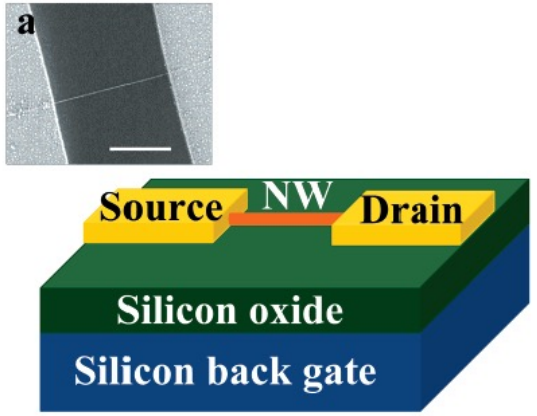
**Reduced dimensionality and high surface/bulk ratio can lead to enhanced and/or novel properties**



D. Saxena et al., *Nat. Phot.* (2013)



P. Yang et al., *Nano Lett.* (2010)




W. Lu and C. M. Lieber, *J. Phys. D* (2006)

## LEDs and microcavity lasers

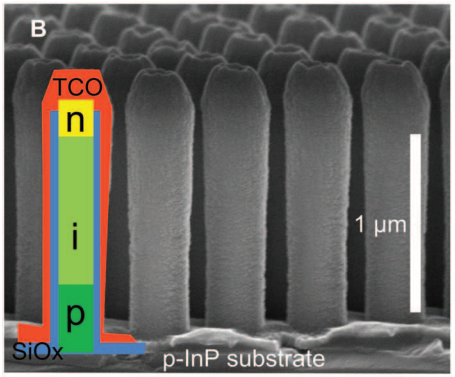
- Nanoscale light sources
- Higher efficiency due to lack of defects

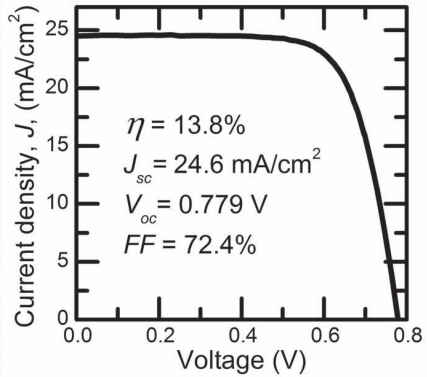
## Interfacing with Cells

- Control signal propagation and communication

## Transistors

- Improved performance
- Small size



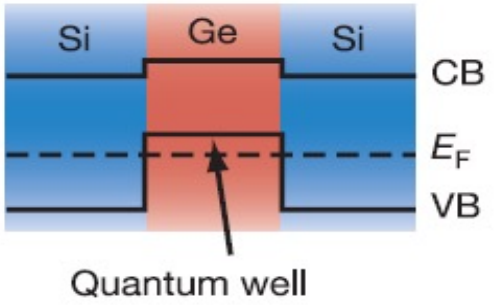

J. Wallentin et al., *Science* (2013)

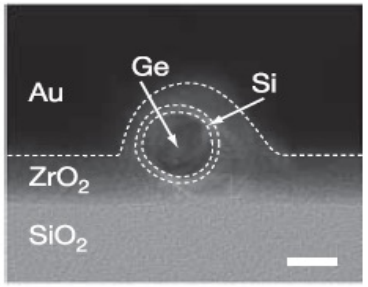
## Photovoltaics

- NW arrays used for light trapping
- Up to 13.8% efficiency for InP-based NW array solar cells

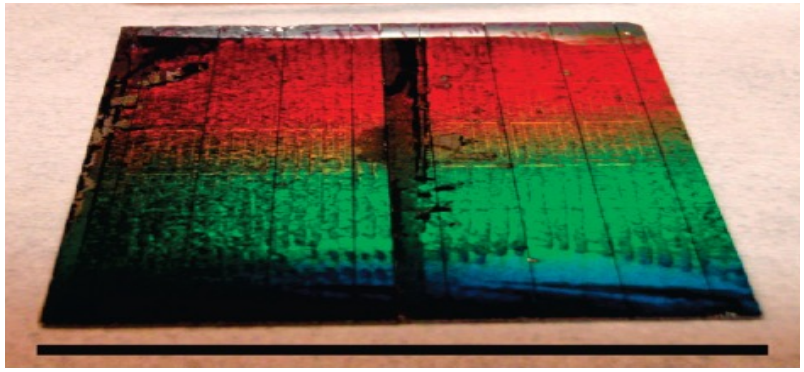
# Semiconductor nanowire heterostructures

**Nanowire (NW) heterostructures have enabled researchers to tailor NW properties for exploring fundamental phenomena and use in a variety of applications**

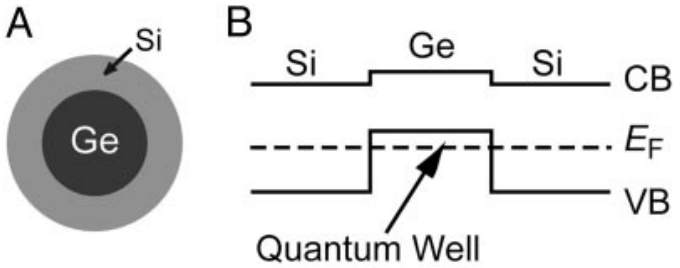


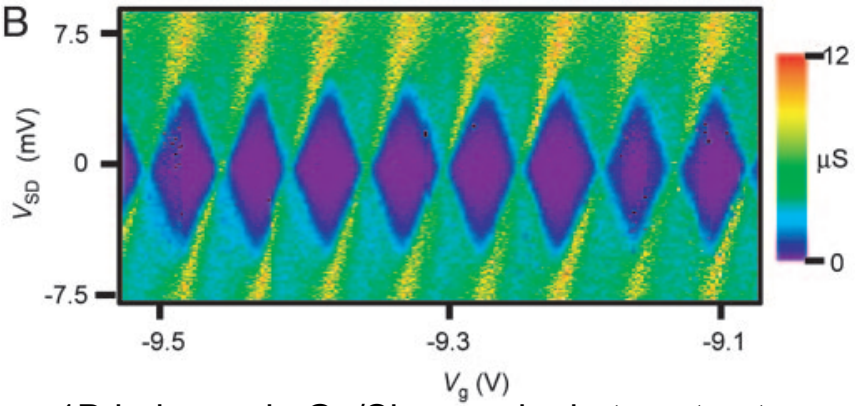




Ge/Si nanowire heterostructures as high-performance field-effect transistors
J. Xiang et al., *Nature* (2006)



Light trapping in silicon nanowire solar cells
E. Garnett & P. Yang, *Nano Lett*. (2010)



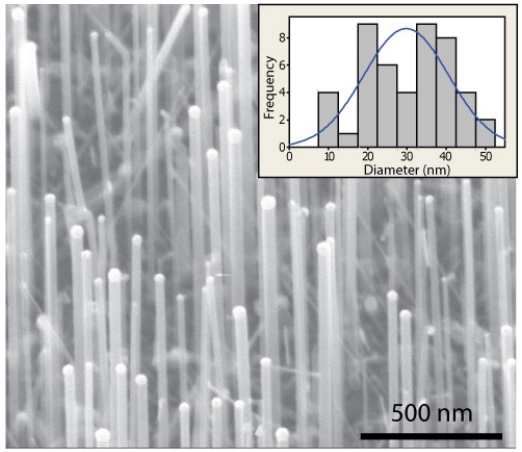




1D hole gas in Ge/Si nanowire heterostructures
W. Lu et al., *PNAS* (2005)

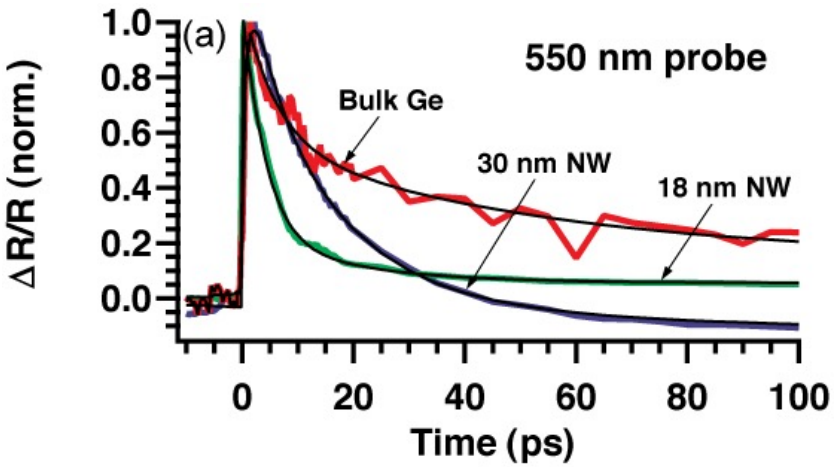
- **Understanding carrier relaxation and transport in NW heterostructures is critical**

# Ultrafast dynamics in NW ensembles

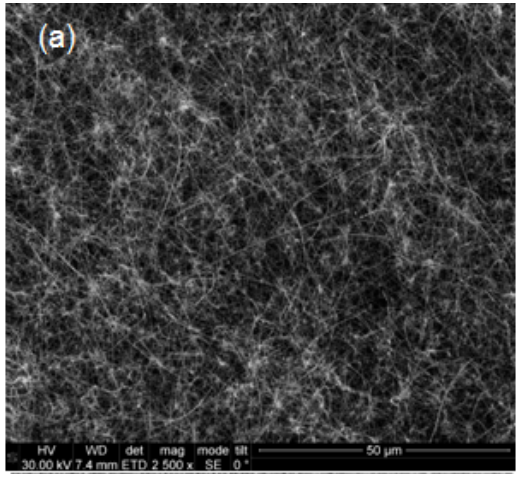
**Ge NWs***

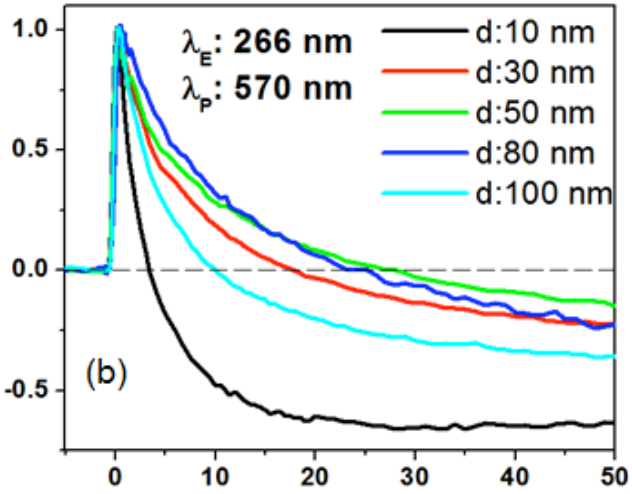



30 nm nanowires




**Si NWs****



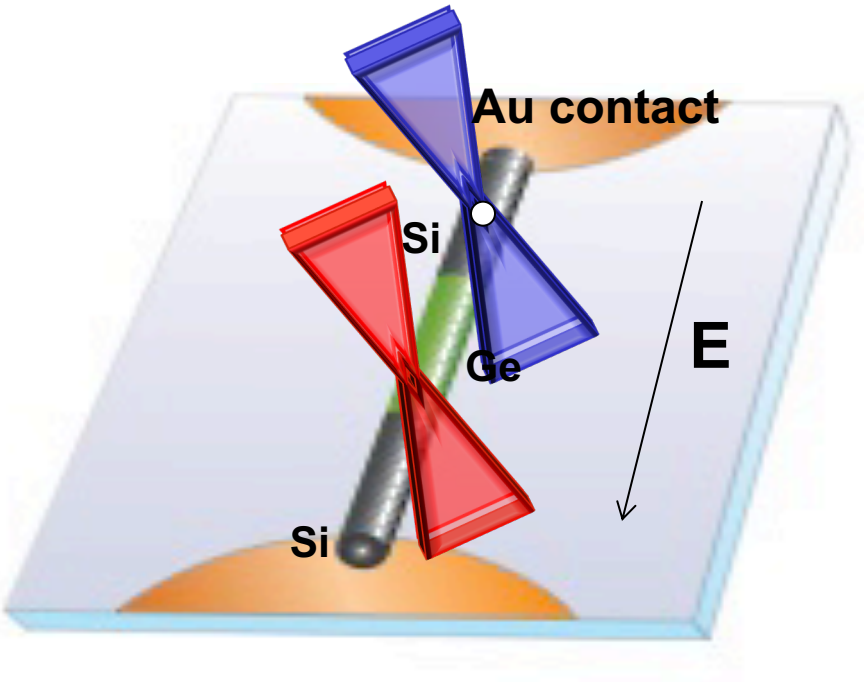




*R. P. Prasankumar et al, *Nano Lett*. **8**, 1619 (2008)

**A. Kar et al., *IEEE Journal of Selected Topics in Quantum Electronics* **17**, 889 (2011)

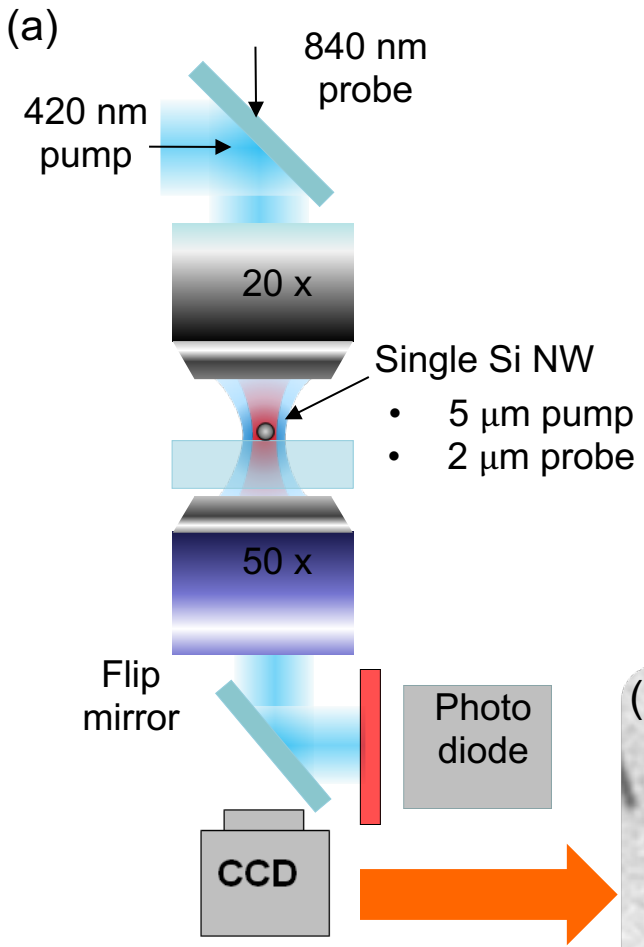
Los Alamos NATIONAL LABORATORY EST.1943

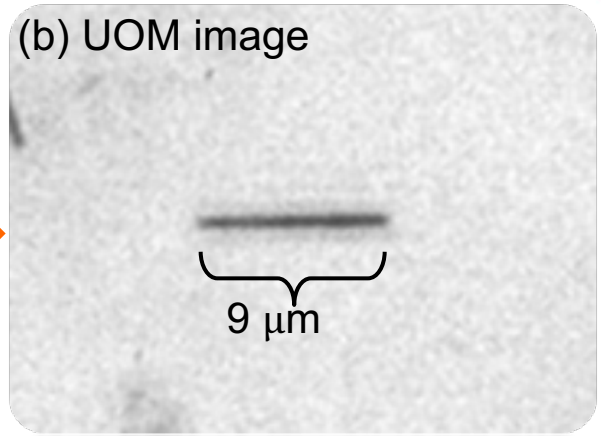
# Mapping carrier dynamics in nanowire heterostructures

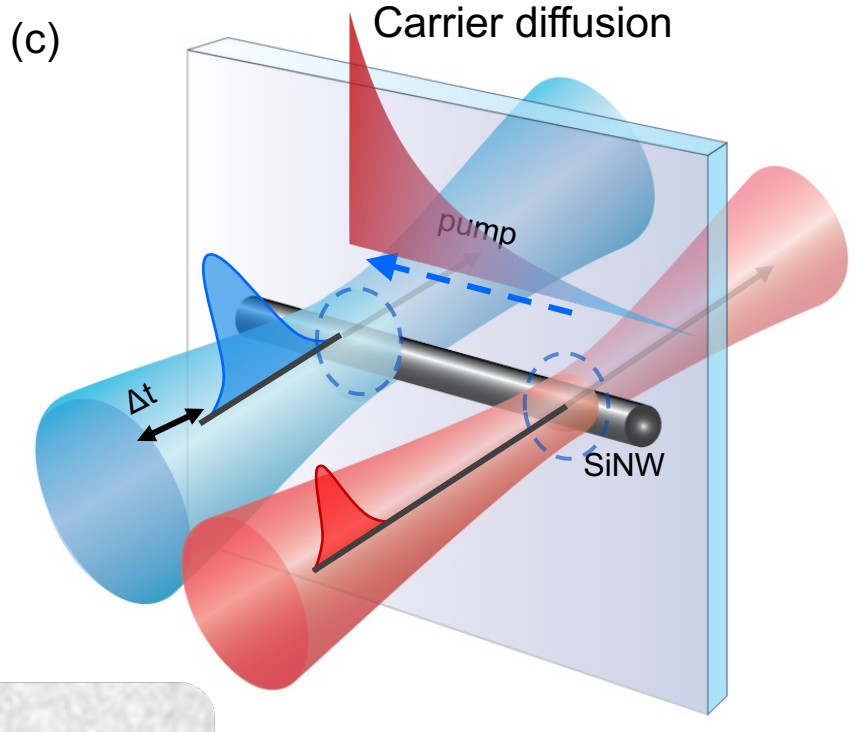



- Ideal experiment: Spatiotemporally map electron and hole distribution throughout individual NWs and NW heterostructures to understand intrinsic optical and electronic properties
- Other experimental techniques (transport, SPCM, TRPL) cannot do this
- Ultrafast optical microscopy (UOM) is an ideal technique for spatiotemporally resolving carrier dynamics in semiconductor NWs
  - Non-contact, high temporal (<100 fs) and spatial (<1 μm) resolution
  - Tune pump and probe photon energies to track energy relaxation

# Ultrafast optical microscopy (UOM)



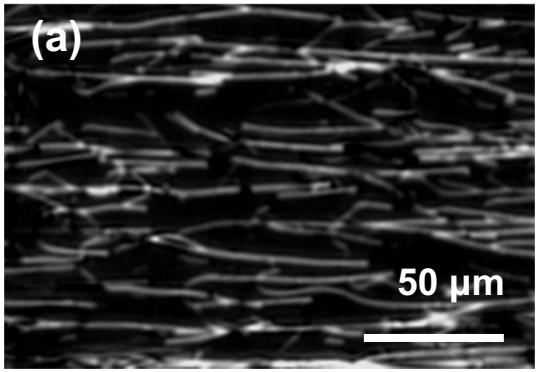



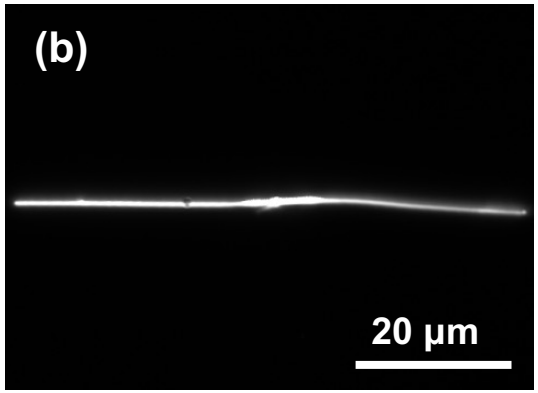



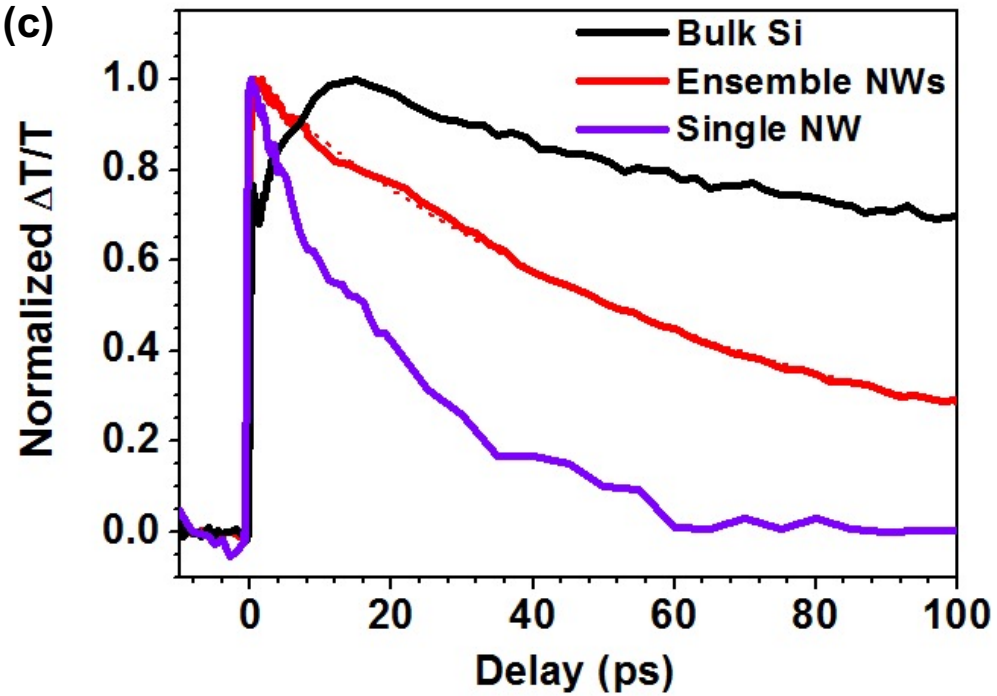

Measured photoinduced change in transmission ΔT/T ~ carrier density

# First ultrafast optical measurements on single Si NWs

Comparing transient carrier dynamics between ensemble and single Si NWs and bulk Si



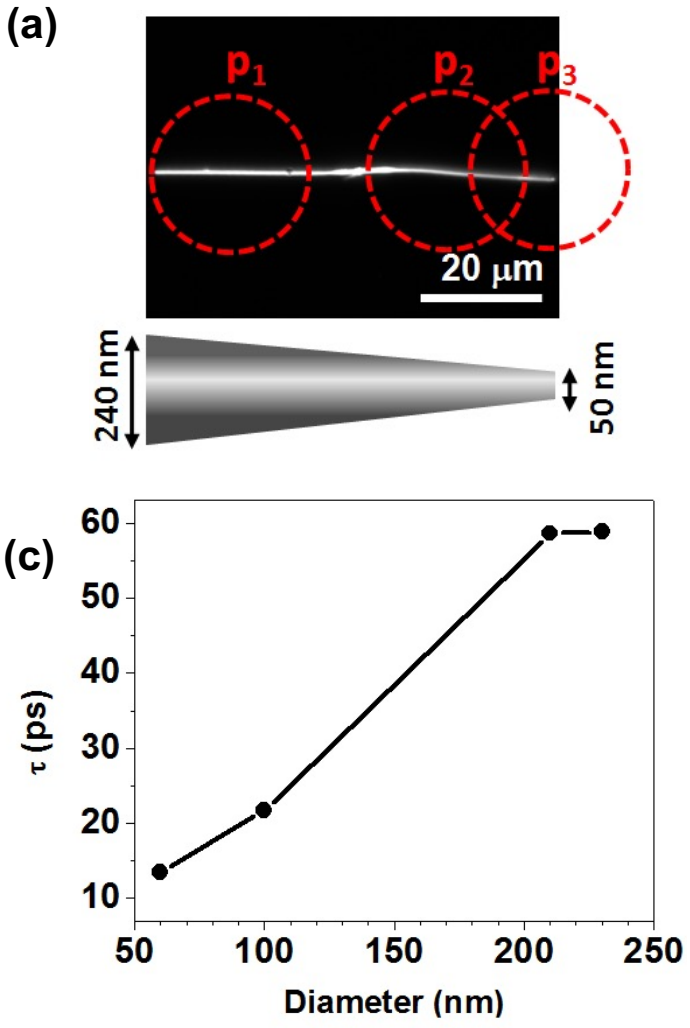



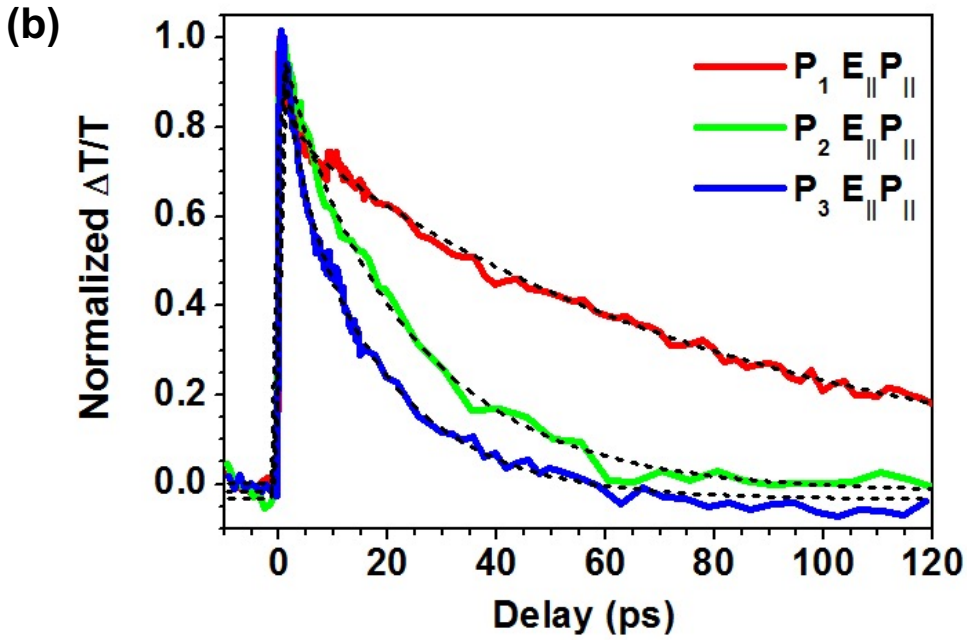

Faster decay in the single NW as compared to bulk Si and NW ensemble



M. A. Seo et al, *Appl. Phys. Lett.* **100**, 071104 (2012)

# Position-dependent dynamics in single Si NWs



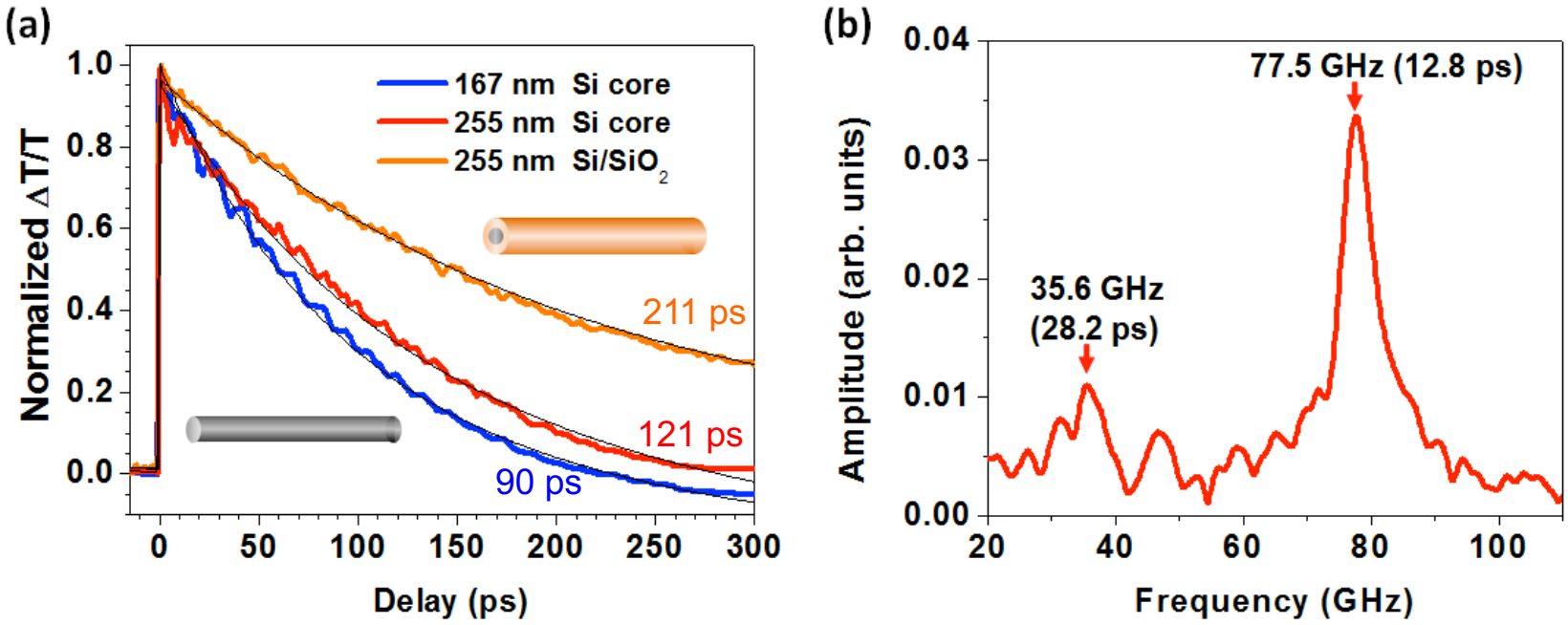

- Time constant τ increases with NW diameter, as in Si NW ensembles
- Surface trapping and recombination influence dynamics
- First observation of this kind in a single NW



M. A. Seo et al, *Appl. Phys. Lett.* **100**, 071104 (2012)

# Ultrafast dynamics in radial Si NW heterostructures

- Photoinduced transmission changes in single Si bare and core-shell NWs
  - Coherent acoustic phonon oscillations observed in both samples




- Propagating sound wave generated through absorption of optical pulse in Si core
  - Main oscillation independent of NW diameter->corresponds to well known Brillouin oscillations
  - Weaker peak likely due to breathing mode as observed by other groups

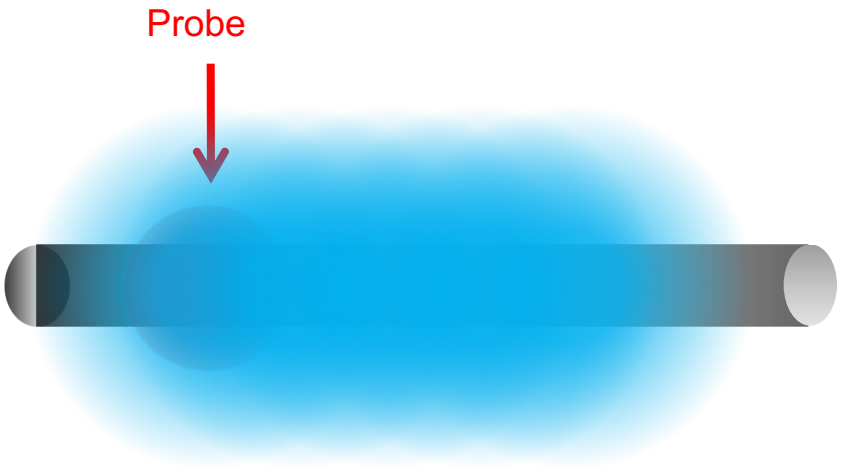


M.A. Seo et al., *Nano Lett.* **12**, 6334 (2012)

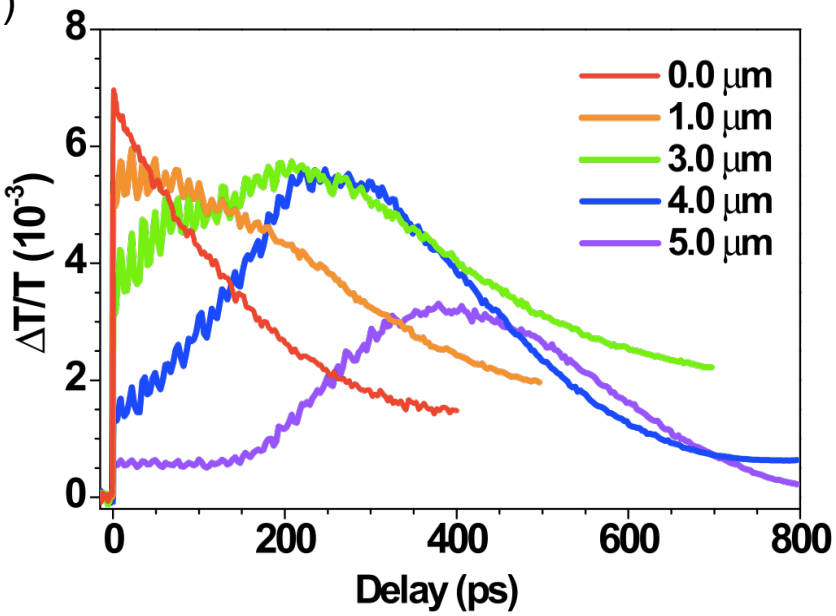
# Space-and-time-dependent charge transport

- UOM measurements on single Si core and Si/$SiO_2$ core-shell NWs
  - Probe beam fixed at one end of the NW, pump beam moved from overlap position to a maximum separation of 5 μm along the NW
  - Rise time increases with pump-probe separation for both bare Si NW and Si/$SiO_2$ NW

(a)




(b)



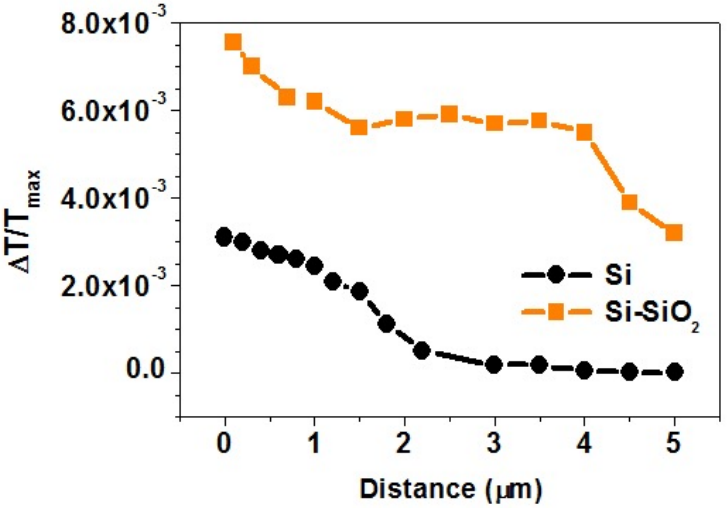

- Direct measurement of charge separation and carrier transport along NWs



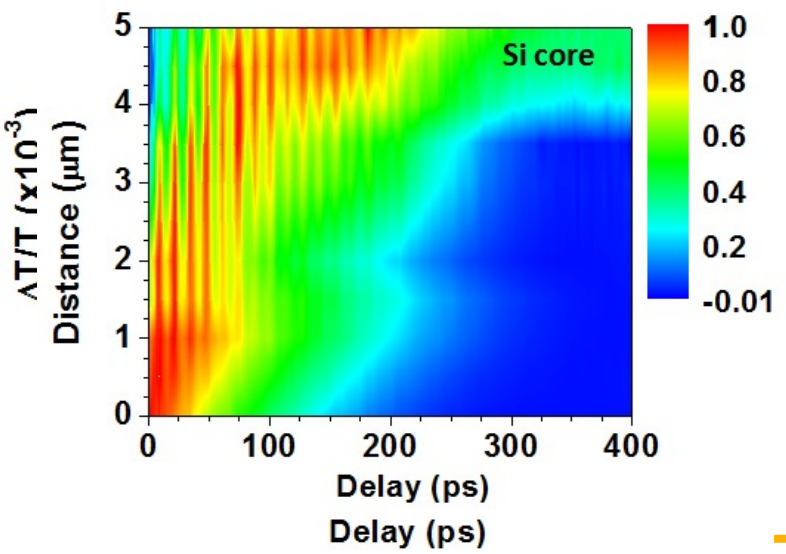
M.A. Seo et al., *Nano Lett.* **12**, 6334 (2012)

# Ultrafast charge transport in single Si NWs

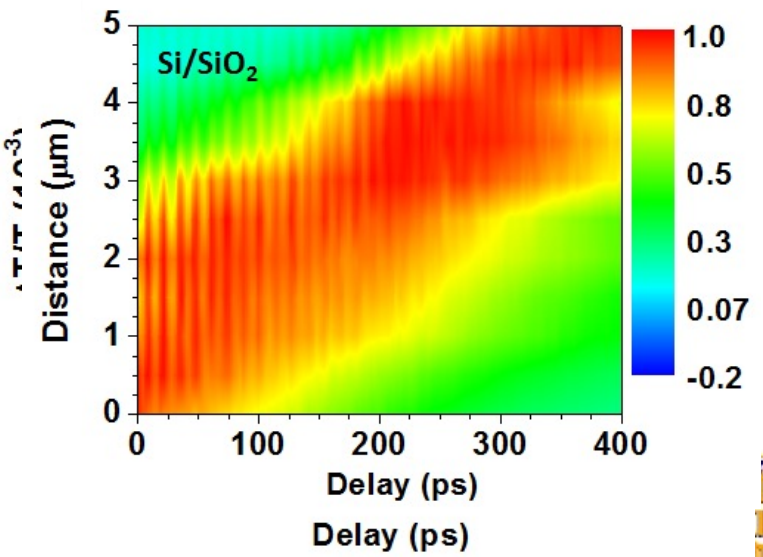
- Photoinduced transmission changes as a function of pump-probe separation
- Maximum ΔT/T signal rapidly decreases for bare Si NW
  - Due to surface trapping and recombination
- Maximum ΔT/T signal persists until 4 μm separation for $Si/SiO_2$ core-shell NW
  - $SiO_2$ shell passivates surface, minimizing trapping and recombination




(a)

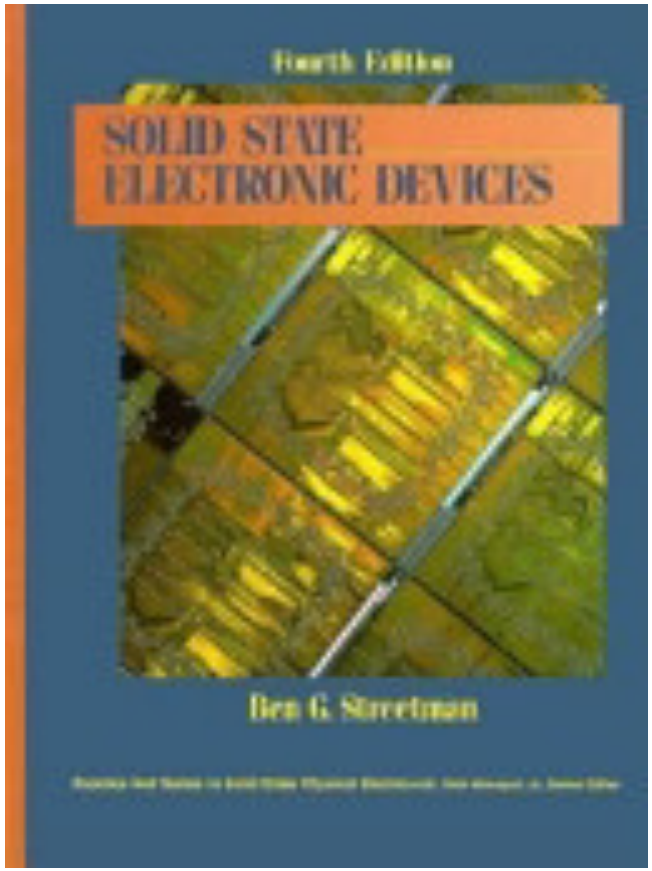



(b)




M.A. Seo et al., *Nano Lett.* **12**, 6334 (2012)

# Carrier diffusion in one dimension



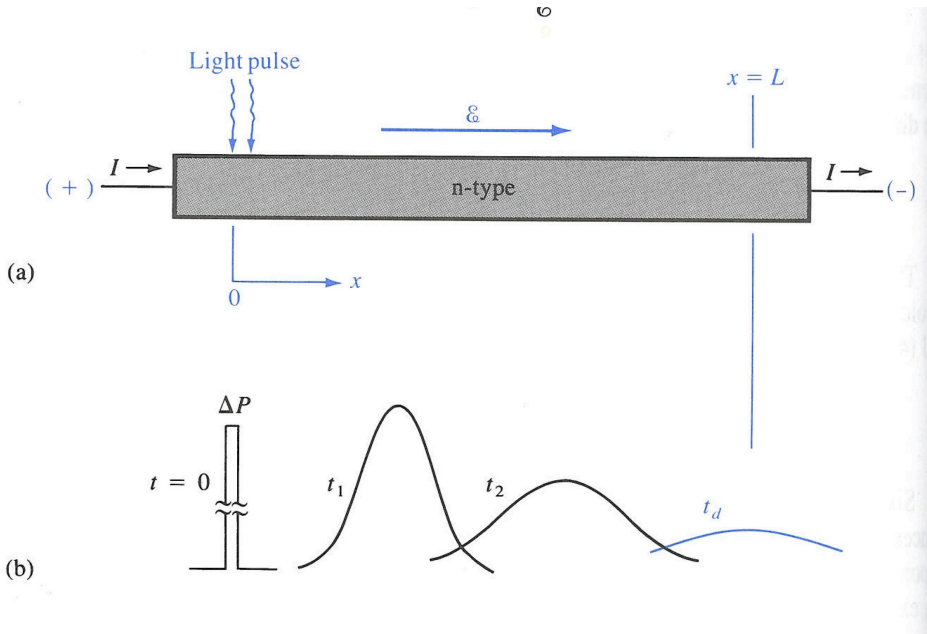




**Figure 4-18** Drift and diffusion of a hole pulse in an n-type bar: (a) sample geometry; (b) position and shape of the pulse for several times during its drift down the bar.

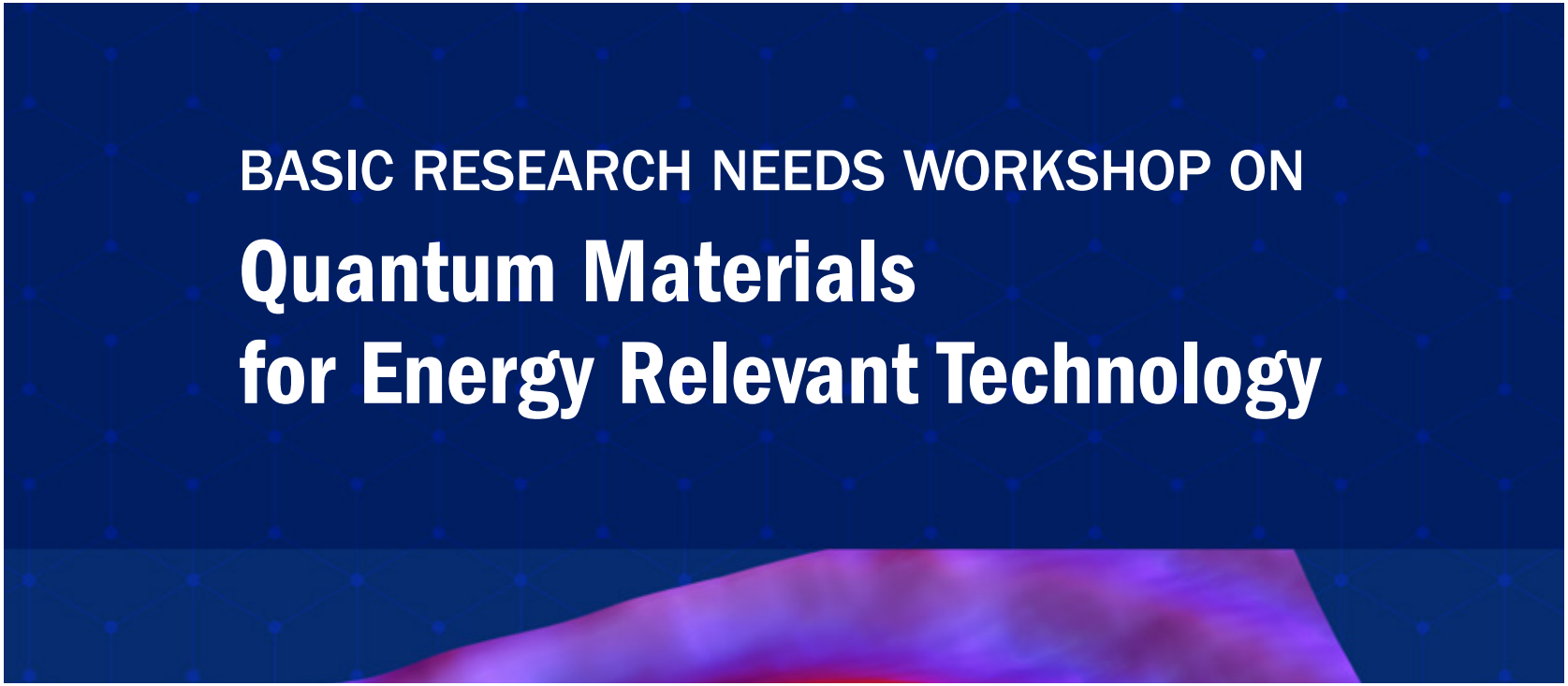
Ultrafast optical microscopy on semiconductor nanowires enables us to study an actual “textbook” case of 1D carrier transport!

# Outline

**We use femtosecond pump-probe spectroscopy to study complex materials, revealing new information about their properties that can impact future applications**

- Background on ultrashort pulse generation and pump-probe spectroscopy
- Examples:
  - Ultrafast carrier relaxation across multiple spatial dimensions in a semiconductor quantum dots-in-a-well (DWELL) heterostructure
    - Can we track carrier relaxation from 3D to 2D to 0D in a novel heterostructure?
  - Time-resolved carrier dynamics in ensemble and individual semiconductor nanowires
    - What pathways do photoexcited carriers take as they lose energy in a quasi-1D system?
- **Going beyond simple semiconductors: using ultrashort light pulses to probe and even control complex quantum materials**

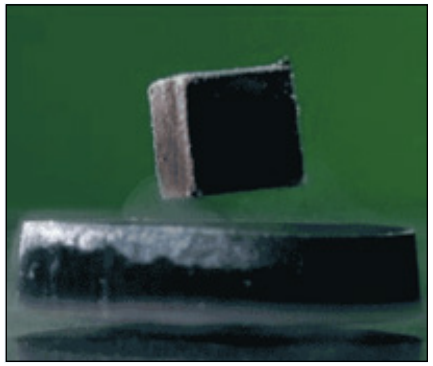
# Going beyond conventional materials



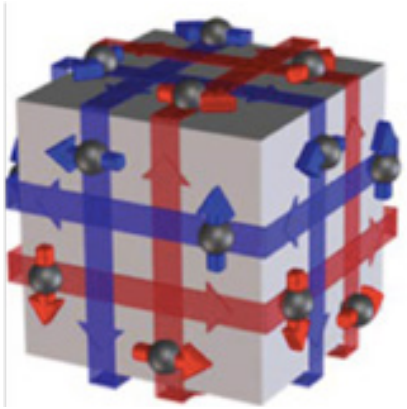

Quantum materials are solids with exotic physical properties, arising from the quantum mechanical properties of their constituent electrons; such materials have great scientific and/or technological potential.

# Different types of quantum materials

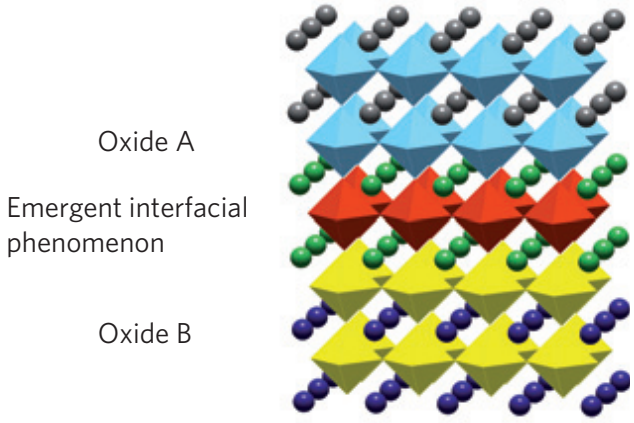
**Strongly correlated electron materials** (superconductors, magnets, multiferroics, quantum spin liquids)

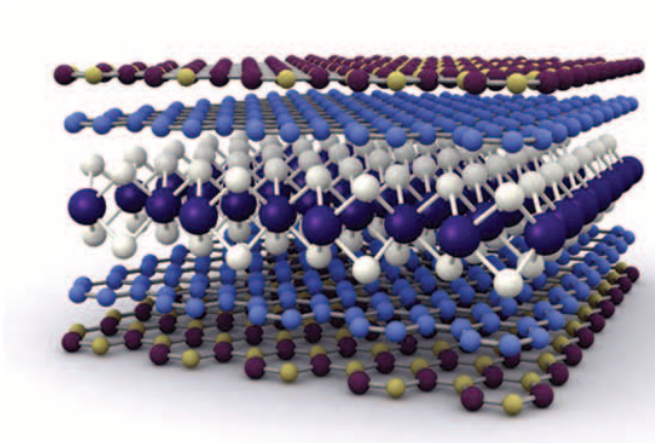


**Dirac materials** (graphene, topological insulators, Weyl/Dirac semimetals)

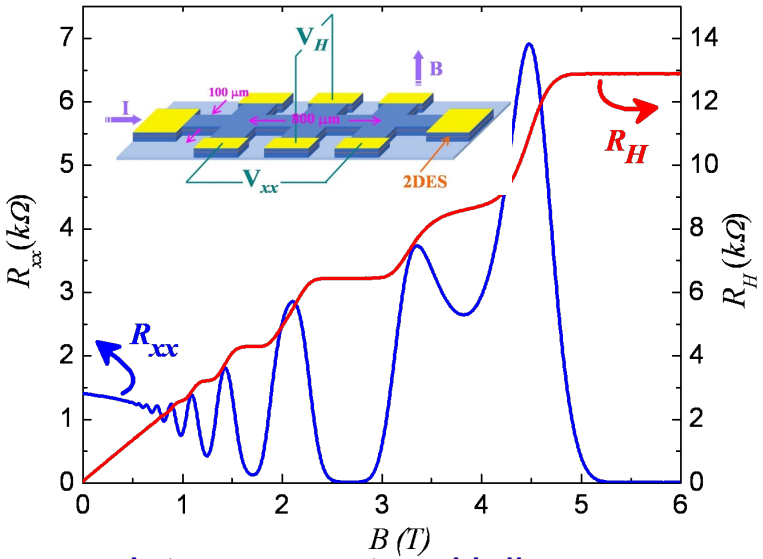


**Heterostructures** (transition metal oxides or dichalcogenides)



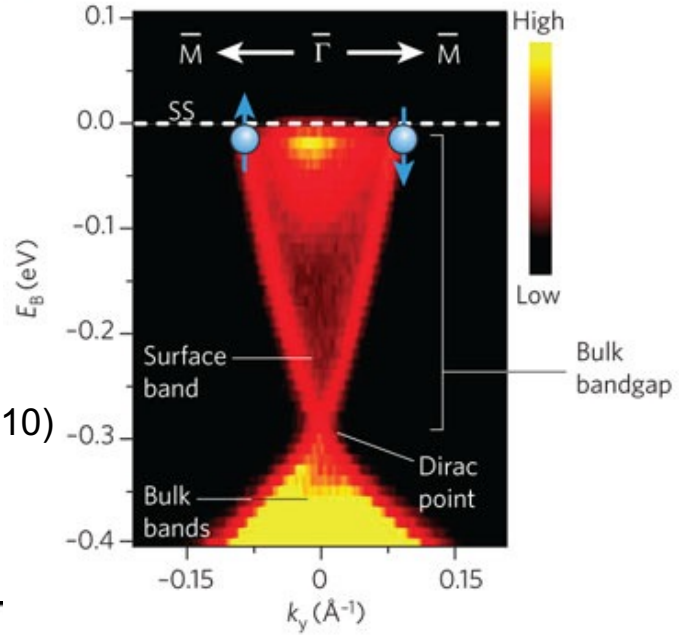
# Novel properties of quantum materials

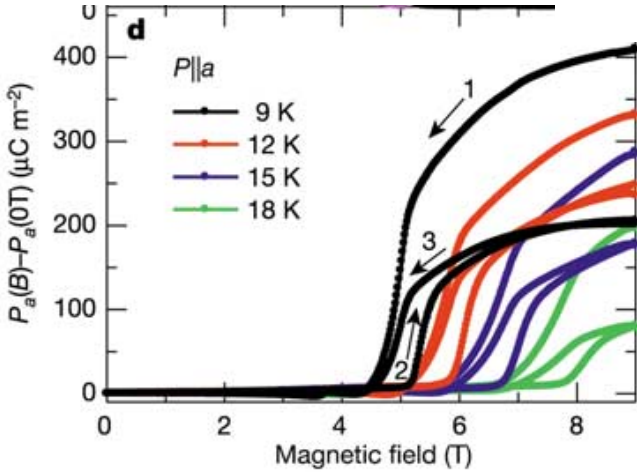



Integer quantum Hall effect in a two-dimensional electron gas (2DEG)

Topological surface state in $Bi_2Se_3$

J. Moore, *Nature* (2010)

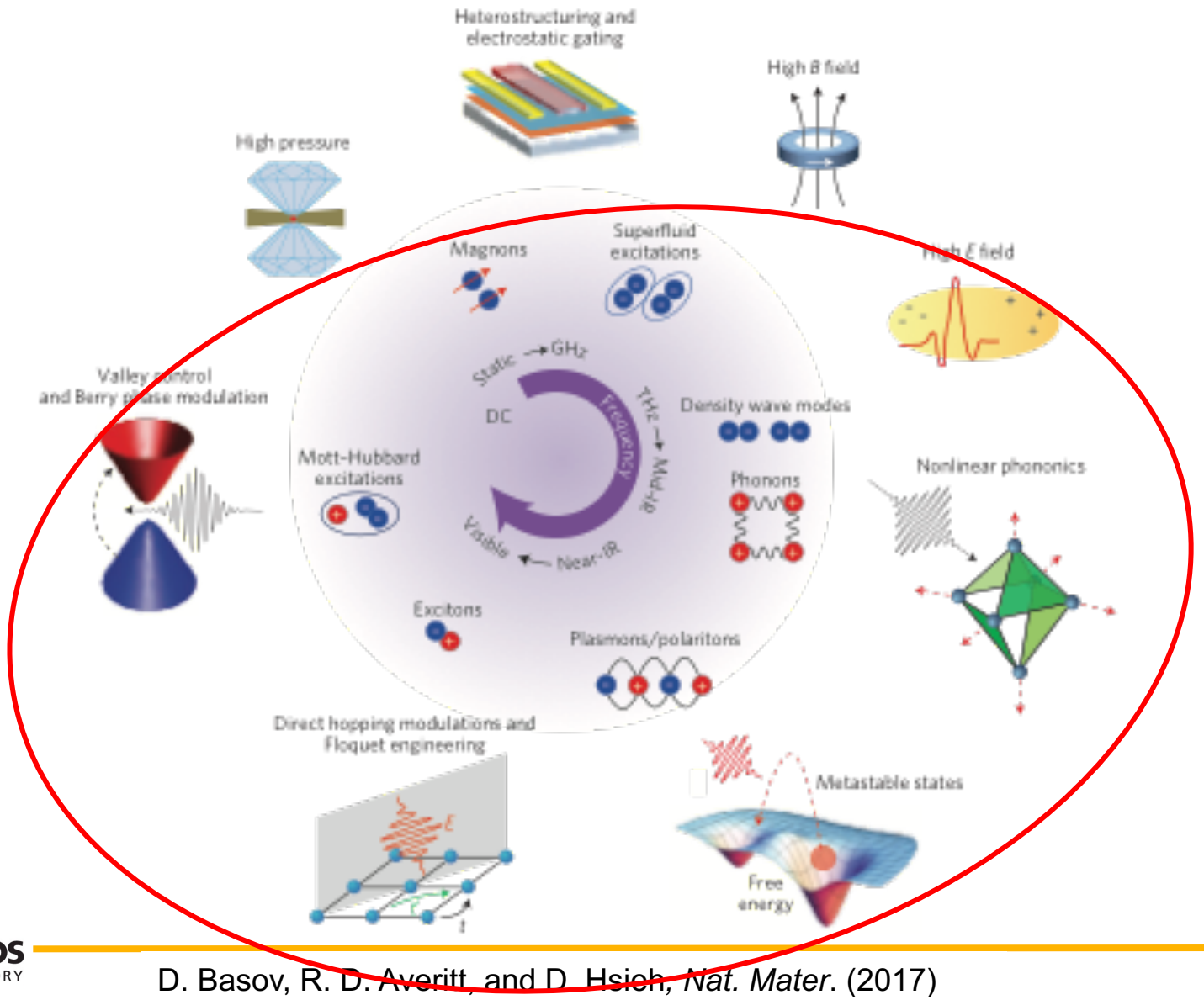



Large magnetoelectric coupling in $TbMnO_3$

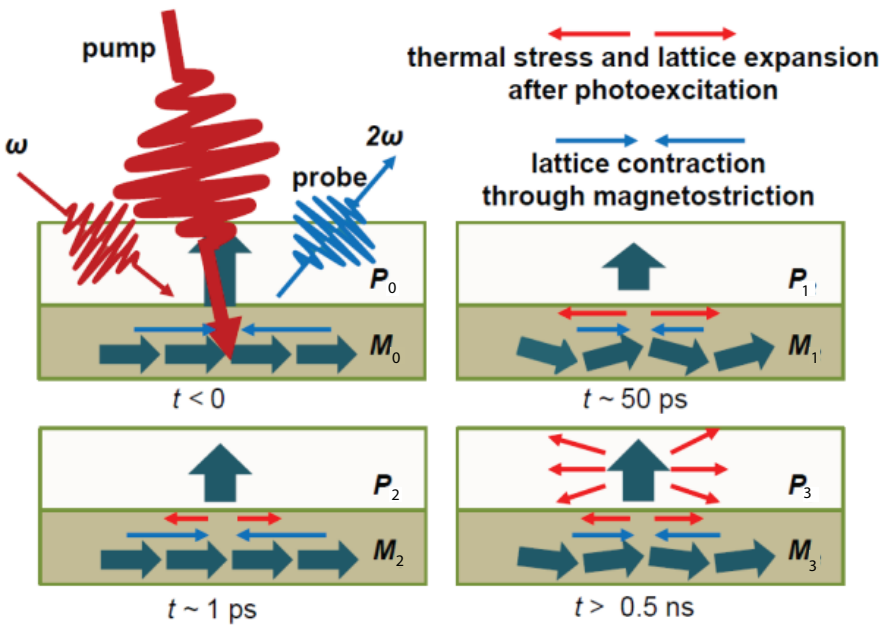
**The allure of quantum materials lies in their potential to provide new functionalities that go beyond those provided by conventional solids**

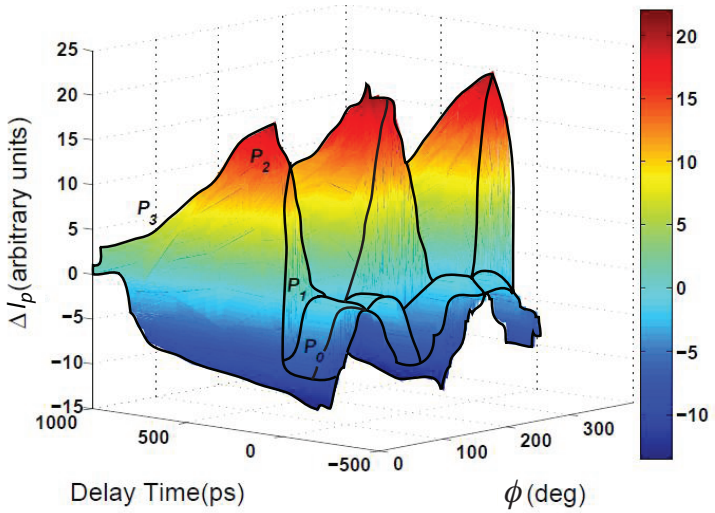
# Studying quantum materials with light




D. Basov, R. D. Averitt, and D. Hsieh, *Nat. Mater*. (2017)

# Using Ultrashort Optical Pulses to Couple Ferroelectric and Ferromagnetic Order in an Oxide Heterostructure




Multiferroic materials, in which ferroelectric (FE) polarization and magnetic order can be coupled, have great potential for applications in data storage and magnetic switching via their intrinsic magnetoelectric (ME) coupling.

– We used femtosecond optical pulses to modify FM order in a $Ba_{0.1}Sr_{0.9}TiO_3$(BSTO) /$La_{0.7}Ca_{0.3}MnO_3$ (LCMO) heterostructure, after which time-resolved second harmonic generation (TR-SHG) was used to study changes in FE polarization

Our experiments revealed that spin-lattice relaxation governs the timescale for ME coupling, which will impact device applications of these oxide heterostructures



Y. M. Sheu, et al, *Nat. Comm.* **5**, 5832 (2014)

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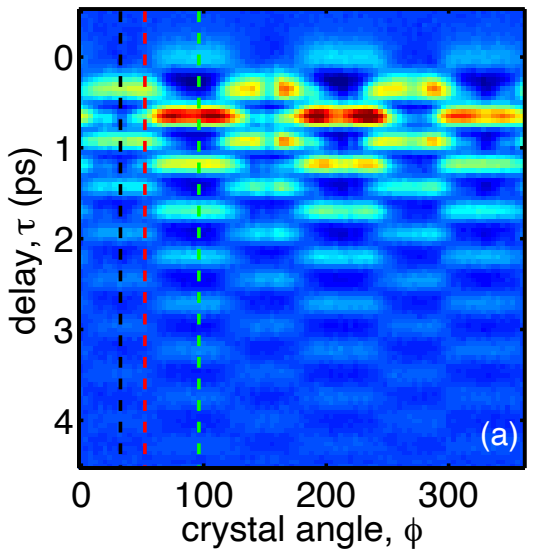
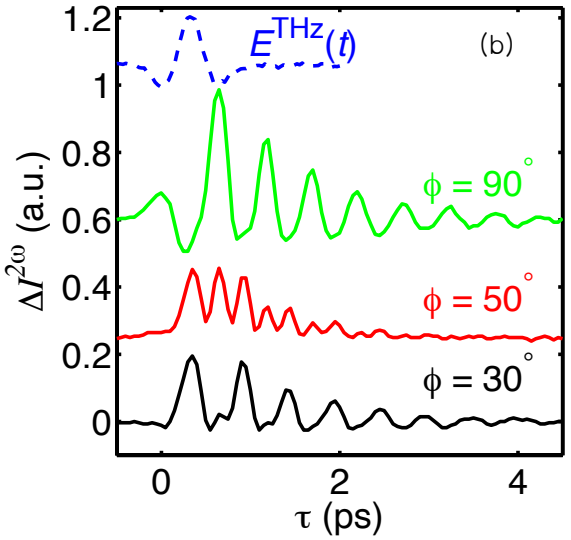
# Probing and Controlling Terahertz-Driven Structural Dynamics with Surface Sensitivity




Topological materials have attracted a lot of attention due to the new phenomena they exhibit, with potential applications in quantum information science.

We used intense THz pulses to excite a phonon mode in the topological insulator $Bi_2Se_3$ and second harmonic generation (SHG) to track the ensuing structural dynamics.

- Intense THz pulses resonantly excited the infrared-active $E_{1u}$ phonon mode in $Bi_2Se_3$
- The resulting dynamic structural changes led to a large modulation of the SHG signal with two oscillatory components: one from the surface, at 1.95 THz, and another from the bulk, at 3.9 THz
- Tuning the time delay between a pair of driving THz pulses enabled us to coherently control the phonon oscillations

This demonstrates a versatile, tabletop tool to probe and control phonon dynamics in nanoscale systems, particularly at surfaces and interfaces.



P. Bowlan, et al, *Optica* **4**, 2334 (2017)

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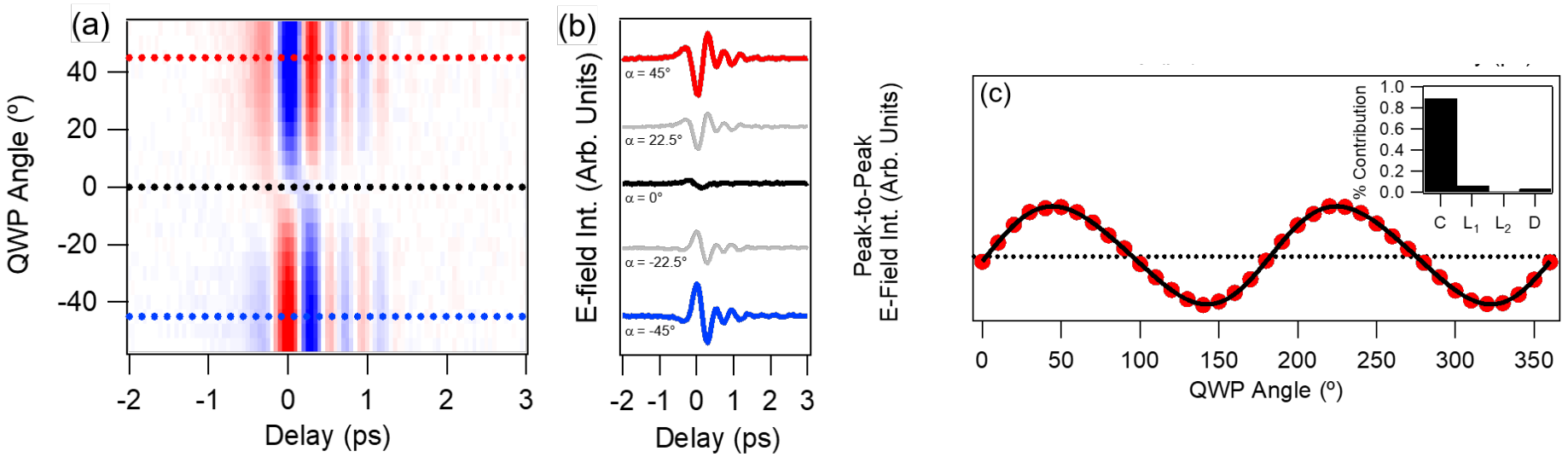
# Tracking Ultrafast Photocurrents in the Weyl Semimetal TaAs Using THz Emission Spectroscopy




Weyl semimetals extend topological phenomena to 3D, resulting in new phenomena that cannot be observed in topological insulators.

We used THz emission spectroscopy to track polarization-dependent ultrafast photocurrents in the Weyl semimetal TaAs, shedding new light on their origin.

- Femtosecond optical pulses with controlled circular polarization were used to drive ultrafast in-plane injection currents and out-of-plane shift currents in TaAs
- By controlling the polarization, we could control the direction of the currents

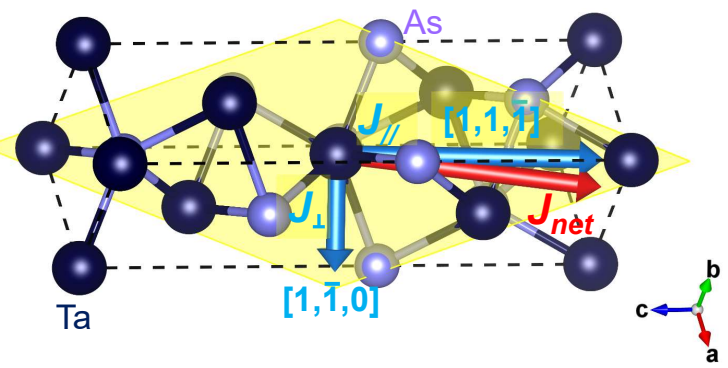
This demonstrates that transient photocurrents in TaAs are inherent to the underlying crystal symmetry of these materials.

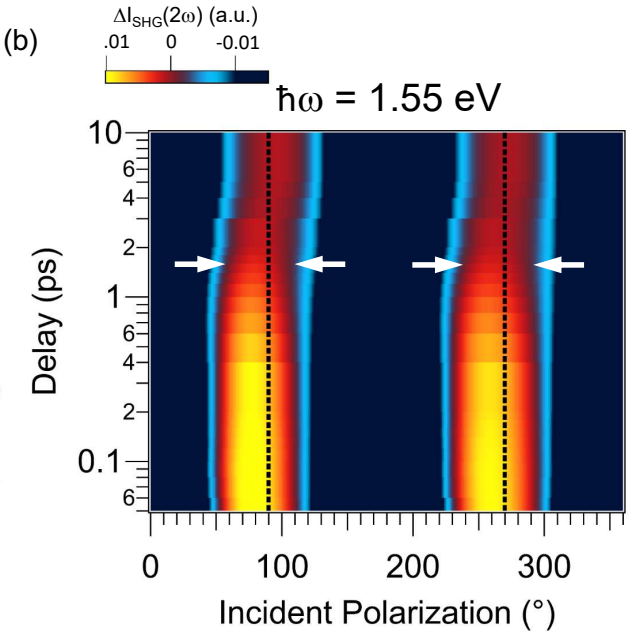


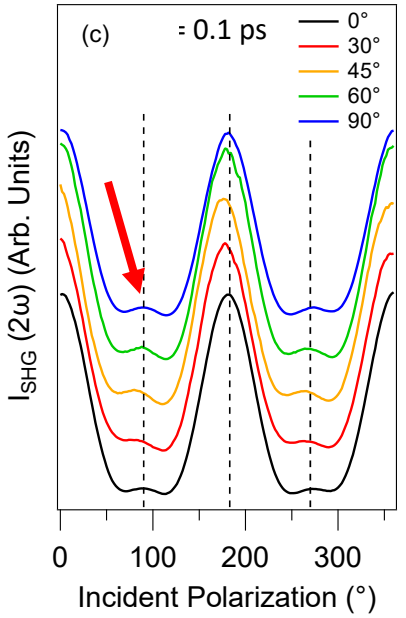
N. Sirica,et al, *Phys. Rev. Lett.* **122**, 197401 (2019)

Los Alamos NATIONAL LABORATORY EST.1943

# Photocurrent-Driven Transient Symmetry Breaking in the Weyl Semimetal TaAs




Using time-resolved SHG, we demonstrated that optically driven photocurrents can break electronic symmetry in TaAs, opening up a new way to control topology.

- We used TR-SHG to optically drive photocurrents and detect the ensuing symmetry changes
- Additional features appear in the SHG pattern due to photocurrent-driven symmetry breaking
- Varying the polarization of the driving pulse enables us to control the degree of symmetry breaking

Our results demonstrate a new way to control the properties of quantum materials by using ultrafast photocurrents to break electronic symmetries, which will be particularly significant for applications of topological materials.



N. Sirica et al, *Nature Materials* (in press) (2021)

# Conclusion

- Ultrafast processes are occurring all around us, all the time, that are completely beyond what we can perceive
- Short (femtosecond) pulses of light are the only way to measure most of these phenomena
- Using femtosecond pulses, we can measure dynamic processes as well as fundamental material properties
  - Ultrafast carrier relaxation from 3D to 2D to 0D in a DWELL heterostructure
  - Tracking carrier relaxation and diffusion in quasi-1D semiconductor nanowires

This is only the tip of the iceberg: new techniques in ultrafast spectroscopy are continually enabling us to look at faster timescales, smaller length scales, and broader ranges of the spectrum, making it possible to “see” more and more phenomena occurring faster than the blink of an eye!